NOTICE: When government or other drawings, specifications or other data are used for any purpose other than in connection with a definitely related government procurement operation, the U. S. Government thereby incurs no responsibility, nor any obligation whatsoever; and the fact that the Government may have formulated, furnished, or in any way supplied the said drawings, specifications, or other data is not to be regarded by implication or otherwise as in any manner licensing the holder or any other person or corporation, or conveying any rights or permission to manufacture, use or sell any patented invention that may in any way be related thereto.

63-4-1

WADD-TR-61-134
Part II

ASTIA 407630

RESEARCH ON WORKABLE REFRACTORY ALLOYS OF TUNGSTEN, TANTALUM, MOLYBDENUM, AND COLUMBIUM

CATALOGED BY DDC
AS AD No.

TECHNICAL DOCUMENTARY REPORT NO. WADD-TR-61-134, PART II

April 1963

Directorate of Materials and Processes
Aeronautical Systems Division
Air Force Systems Command
Wright-Patterson Air Force Base, Ohio

Project No. 7351, Task No. 735101

(Prepared under Contract No. AF33(616)-8135 by Crucible Steel Company of America, Pittsburgh, Pennsylvania; R. C. Westgren, V. R. Thompson, and V. C. Petersen, authors)

DDC
RELEASED
JUN 25 1963
TISIA D

NOTICES

When Government drawings, specifications, or other data are used for any purpose other than in connection with a definitely related Government procurement operation, the United States Government thereby incurs no responsibility nor any obligation whatsoever; and the fact that the Government may have formulated, furnished, or in any way supplied the said drawings, specifications, or other data, is not to be regarded by implication or otherwise as in any manner licensing the holder or any other person or corporation, or conveying any rights or permission to manufacture, use, or sell any patented invention that may in any way be related thereto.

Qualified requesters may obtain copies of this report from the Armed Services Technical Information Agency, (ASTIA), Arlington Hall Station, Arlington 12, Virginia.

This report has been released to the Office of Technical Services, U.S. Department of Commerce, Washington 25, D.C., in stock quantities for sale to the general public.

Copies of this report should not be returned to the Aeronautical Systems Division unless return is required by security considerations, contractual obligations, or notice on a specific document.

B

31-764C, 600, 6-14-63

FOREWORD

This report was prepared by the Crucible Steel Company of America, Pittsburgh, Pennsylvania, under U. S. Air Force Contract No. AF33(616)-8135. The contract was initiated under Project No. 7351, "Metallic Materials," Task No. 735101, "Refractory Metals." The work was administered under the direction of the Directorate of Materials and Processes, Aeronautical Systems Division, with James T. Gow serving as project engineer.

This report covers work conducted from April 15, 1961 to November 15, 1962.

ABSTRACT

Under a previous contract, the W-Ta-Mo-Cb alloy system was investigated, and several tungsten- and tantalum-rich alloys were developed and evaluated in the form of extruded bars. Many of these alloys exhibited very high strengths at 3000 F; in fact, the tensile strengths of some alloys were in excess of 60,000 psi. The present work was a continuation of this effort and was aimed at producing and evaluating alloys from the W-Ta-Mo-Cb system in the form of sheet (Phase I) and increasing the high-temperature strength of alloys of this type by the formation of dispersed carbides (Phase II).

For Phase I, small cylindrical ingots of six selected alloys and unalloyed tungsten were consumably vacuum arc-melted by a multiple electrode technique and successfully extruded to sheet bars. Two alloys and unalloyed tungsten were rolled to sheet and recrystallization temperatures, bend transition temperatures, and high-temperature tensile properties were determined. The other four alloys could not be rolled to sheet by the techniques attempted in this program.

Experiments under Phase II resulted in the development of a successful technique for consumably vacuum arc melting 88W-12Cb alloys that contained small amounts of vanadium, zirconium, and carbon. In the as-extruded condition, the alloys showed 3500 F tensile strengths of 49,000 to 57,000 psi—this constituted a twofold increase in strength above that of the base solid solution composition.

This technical report has been reviewed and is approved.

I. Perlmutter

I. PERLMUTTER
Chief, Physical Metallurgy Branch
Metals and Ceramics Laboratory
Directorate of Materials & Processes

TABLE OF CONTENTS

	PAGE
INTRODUCTION.	1
PHASE I	2
Alloy Selection.	2
Melting Experiments.	3
Extrusion Experiments.	8
Rolling Experiments.	11
Metallographic Preparation	21
Recrystallization Studies.	22
Bend Transition Studies.	24
Tension Tests.	26
Summary.	27
PHASE II.	30
Melting Experiments.	30
Extrusion Experiments.	34
Metallographic Studies	34
Mechanical Tests	36
Summary.	37

LIST OF TABLES

		PAGE
Table I	Tensile Properties at 3000 F of As-Extruded Materials.	39
Table II	Analyses of Electrode Rods	40
Table III	Results of Hardness Tests on Consumable-Electrode Vacuum-Arc-Melted Ingots for Phase I.	41
Table IV	Results of Phase I Extrusion Experiments . .	43
Table V	Rolling Schedules for Reducing Sheet Bars to Intermediate-Gauge Sheets	46
Table VI	Rolling Schedules for Reducing Intermediate-Gauge Sheets to Final-Gauge Sheets	47
Table VII	Results of Rolling Experiments on the 75Ta-25W Alloy	48
Table VIII	Results of Rolling Experiments on the 88Ta-12Mo Alloy and Unalloyed Tungsten . . .	49
Table IX	Summary of Melting, Extrusion, and Rolling for Phase I.	50
Table X	Results of Recrystallization Studies on the 75Ta-25W Alloy	51
Table XI	Results of Recrystallization Studies on the 88Ta-12Mo Alloy.	52
Table XII	Results of Recrystallization Studies on Unalloyed Tungsten	53
Table XIII	Summary of Recrystallization Temperatures of Selected Warm-Worked Refractory Metals and Alloys.	54
Table XIV	4T Bend Transition Data on Sheet Material. .	55
Table XV	Results of Vacuum Tension Tests on Sheet Specimens of 75Ta-25W, 88Ta-12Mo, and Unalloyed Tungsten	56

LIST OF TABLES (continued)

		PAGE
Table XVI	Results of Phase II Extrusion Experiments.	57
Table XVII	As-Extruded Tensile Properties of Phase II Alloys in Vacuum.	58

LIST OF FIGURES

		PAGE
Figure 1	As-Extruded Tensile Strength at 3000 F as a Function of Composition. . . .	59
Figure 2	Over-All View of the Melting Equipment.	60
Figure 3	Ingots of the 68Ta-20W-12Mo Alloy.	61
Figure 4	Ingots of the 68W-20Ta-12Mo and the 68Ta-20W-12Mo Alloys after Lathe Turning and Cylindrically Grinding	62
Figure 5	Typical Extrusion Billets and Die Inserts.	63
Figure 6	Typical Sheet-Bar Extrusions	64
Figure 7	Sheet-Bar Extrusions with Reductions of Area of 67, 70, and 76%.	65
Figure 8	Representative As-Extruded Microstructures of the 75Ta-25W and 88Ta-12Mo Alloys . . .	66
Figure 9	Representative As-Extruded Microstructures of the 68W-20Ta-12Mo and 68Ta-20W-12Mo Alloys	67
Figure 10	Representative As-Extruded Microstructures of the 44W-44Ta-12Cb and 88W-6Mo-6Cb Alloys	68
Figure 11	Zirconia-Coated Rolls used in the Stanat Mill.	69
Figure 12	Setup for Plasma Heating and Rolling . . .	70
Figure 13	75Ta-25W Alloy (CEVA Heat 165) after 69% Reduction in Thickness by Hot Rolling. . .	71
Figure 14	Sheet Bars Hot-Rolled to Intermediate-Gauge Sheet.	72
Figure 15	Sheet Samples of the 75Ta-25W Alloy. . . .	73

LIST OF FIGURES (continued)

PAGE

Figure 16 Microstructure of a Sheet-Bar Sample of the 44W-44Ta-12Cb Alloy (Heat 173T) that Failed during Hot Rolling. 74

Figure 17 Microstructures of Intermediate- and Final-Gauge Sheets of the 75Ta-25W Alloy. . 75

Figure 18 Microstructures of Intermediate- and Final-Gauge Sheets of the 88Ta-12Mo Alloy . 76

Figure 19 Recrystallization Behavior of the 75Ta-25W Alloy. 77

Figure 20 Recrystallization Behavior of the 88Ta-12Mo Alloy 78

Figure 21 Recrystallization Behavior of Unalloyed Tungsten. 79

Figure 22 Bend Test Specimens 80

Figure 23 Bend Test Equipment 81

Figure 24 4T Bend Transition Curve for Unalloyed Tungsten Sheet after Stress Relief. 82

Figure 25 4T Bend Transition Curve for 88Ta-12Mo Sheet after Stress Relief 83

Figure 26 4T Bend Transition Curve for 75Ta-25W Sheet after Stress Relief 84

Figure 27 4T Bend Transition Curves for Stress-Relieved Sheet. 85

Figure 28 Microtensile Test Specimens 86

Figure 29 As-Cast Microstructure of the W-Ti-Zr-C Alloy (CEVA Heat 205) 87

Figure 30 As-Cast Microstructure of the W-Cb-Zr-C Alloy (CEVA Heat 206) 88

Figure 31 Round-Bar Extrusions of the 88W-12Cb Alloy 89

LIST OF FIGURES (continued)

		PAGE
Figure 32	Round-Bar Extrusions of the Phase II Alloys.	90
Figure 33	Schematic Drawing of Vacuum Heat-Treating Apparatus	91
Figure 34	As-Cast Microstructures of Phase II Alloys.	92
Figure 35	As-Extruded and Solution-Annealed Microstructures of the 88W-12Cb Alloy . . .	93
Figure 36	As-Extruded and Solution-Annealed Microstructures of the W-12Cb-Zr-C Alloy	94
Figure 37	As-Extruded and Solution-Annealed Microstructures of the W-12Cb-V-Zr-C Alloy	95
Figure 38	Tension Test Specimen	96
Figure 39	Tension Specimens Tested at 3500 F.	97
Figure 40	Microstructures of Undeformed and Fractured Portions of Tensile Specimens . .	98

INTRODUCTION

This research program is a continuation of Crucible's previous work on the development of W-Ta-rich alloys from the W-Ta-Mo-Cb alloy system. Under Contract AF33(616)-6172 twenty compositions (unalloyed metals and binary, ternary, and quaternary alloys) were studied, and the following accomplishments resulted:

(1) A melting unit and procedure were developed to successfully melt small cylindrical ingots so that the alloy development program could be conducted expeditiously.

(2) Significant information on the working of W-Ta-base alloys was obtained by high-speed extrusion experiments.

(3) Special testing equipment was designed and constructed to evaluate the properties of refractory alloys at 3000 F and higher.

(4) Significant effects of alloying within the system W-Ta-Mo-Cb, where complete solid solubility exists, were determined.

The two principal objectives of the experimental program conducted under Contract AF33(616)-8135 were as follows:

Phase I: To produce, process to experimental sheet form, and evaluate six alloys selected from those studied under Contract AF33(616)-6172[1];

Phase II: To achieve a further significant increase in strength at 3000 F and higher by formation of dispersed carbides.

As indicated above, this program was divided into two phases which were, to some extent, pursued concurrently.

---

[1] "Research on Workable Alloys of Tungsten, Tantalum, Molybdenum and Columbium," WADD Technical Report 61-134, April 1, 1961.

Manuscript released by authors March 1, 1963 for publication as an ASD Technical Report.

PHASE I

Alloy Selection

The following twenty metals and alloys in the W-Ta-Mo-Cb system were successfully melted and extruded under Contract AF33(616)-6172:

W	Ta	Mo	Cb	W	Ta	Mo	Cb
100	--	--	--	88	--	12	--
75	25	--	--	68	20	12	--
50	50	--	--	44	44	12	--
25	75	--	--	20	68	12	--
--	100	--	--	--	88	12	--
88	--	--	12	88	--	6	6
68	20	--	12	68	20	6	6
44	44	--	12	44	44	6	6
20	68	--	12	20	68	6	6
--	88	--	12	--	88	6	6

The terms of the present contract stated that six alloys should be selected from the twenty alloys studied under Contract AF33(616)-6172[1] on the basis of both strength and fabricability considerations. Since not all of the twenty alloys had been tension-tested at 3000 F because of failures during the machining of test specimens, additional samples of some of the twenty alloys were prepared and tested to provide a complete set of tensile properties upon which the selection of the Phase I alloys could be based.

The melting, extrusion, and testing procedures for the additional samples were the same as those employed under Contract AF33(616)-6172.[1] Enough melting stock was available from the previous contract for preparation of these alloys. The results of the vacuum tension tests conducted at 3000 F on specimens in the as-extruded condition (reduction ratio of approximately 3 to 1) are given in Table I together with previously determined results. The 3000 F tensile strengths of the twenty alloys are also shown graphically in Figure 1.

It is important to note that four alloys of Crucible's refractory alloy development program have exhibited tensile strengths in excess of 60,000 psi at 3000 F. The highest strength, namely, 67,000 psi, was obtained with the 68W-20Ta-12Mo (Alloy 12). In addition, the agreement between the results of duplicate tests has been remarkably good. For example, specimens from different heats of the 68Ta-20W-12Mo alloy (Alloy 14) exhibited tensile strengths of 52,000 and 55,000 psi at 3000 F.

Since previous work had indicated that increased high-temperature strength led to greater difficulties in extrusion, high-strength alloys (greater than 50,000 psi tensile strength at 3000 F) as well as intermediate-strength alloys (25,000 to 50,000 psi tensile strength at 3000 F) were selected for this program to provide varying degrees of fabricability. Alloys with 3000 F tensile strength less than 25,000 psi were not considered to be of interest.

The six alloys approved by ASD for study under Phase I and their as-extruded tensile properties at 3000 F are as follows:

Alloy	Tensile Strength (1000 psi)	Elongation in 0.5 Inch (%)	Reduction of Area (%)
88Ta-12Mo	29	29	24
75Ta-25W	37	3	1
68Ta-20W-12Mo	55	24	40
88W-6Mo-6Cb	62	~6	--
68W-20Ta-12Mo	67	16	23
44W-44Ta-12Cb	54	7	13

Melting Experiments

A consumable-electrode vacuum-arc melting furnace, constructed at this Laboratory for work under Contract No. AF33(616)-6172, was used for melting cylindrical ingots (1-1/4 inches in diameter) of refractory metal alloys. To produce internally sound ingots with good surface quality and low interstitial impurity content, the principles of

consumable-electrode vacuum-arc melting and those of consumable-electrode arc welding were applied in the design and operation of the furnace.

The consumable-electrode process of arc melting was chosen to achieve two prerequisites of good ingot quality. First, continuous melting under stable arc voltage and stable arc length was necessary to obtain internally sound ingots free of surface voids or cold shuts. Second, high-purity melting stock was necessary to obtain alloy ingots without interstitial element contamination. Such stock was available in the form of clean, wrought rods. The unique features of the furnace are its provisions for continuously melting electrodes composed of high-purity wrought rods of tungsten, tantalum, molybdenum, and columbium.

An over-all view of the specially constructed consumable-electrode vacuum-arc melting furnace is shown in Figure 2.

The melting chamber of the furnace consists of a 12-inch-diameter, 14-inch-high Type 304 stainless steel cylinder surrounded by a spiral-flow water jacket. Above the melting chamber is a stainless steel housing that contains Pyrex ports for observation of the arc, electrode feed rolls, and electrical contact assembly for the electrode. The uppermost part of the furnace is the electrode housing which supports and guides the electrodes during melting.

One of the important features of the furnace is the electrode feed mechanism. Electrode feed is accomplished by an arc-voltage controlled motor-gear drive unit which turns a set of feed rolls through a double "O" ring seal in the electrode housing. The largest of the multiple electrode rods is gripped between a narrow, grooved drive roll and a smooth, spring-loaded idling roll. The smaller rods pass between or at the side of the rolls and are not gripped by the rolls. The smaller rods are supported by a clamp attached to the largest rod at the upper end. To ensure traction between the drive roll and the largest of the electrode rods, a serrated, grooved hardened steel roll is used for a tantalum rod and a soft aluminum, rubber-faced roll is used for a tungsten rod.

Electrode feed rate and arc length are automatically controlled at a selected level by application of the arc voltage—reference voltage differential to the field of the drive motor. Both arc voltage and arc current are continuously recorded on an Esterline-Angus recording voltmeter and ammeter during the melting sequence.

Electrical power is passed into the electrode rods by a special sliding contact assembly. A hollow, water-cooled copper block is centrally located 4 inches above the top of the ingot mold. It is connected to the power supply by a copper bar which enters the melting chamber through a combination vacuum seal—electrical insulator made of Teflon. A V-shaped tungsten insert, against which the electrodes slide, is placed in the hollow copper block. A spring mechanism, located on top of the copper block, holds the electrodes in the V-groove ensuring positive electrical contact. The electrode configuration and small size prevented the use of the electrical contact systems common to arc furnaces. The system devised for the present arc furnace has provided uniform, uninterrupted melting of electrodes composed of one to four rods of various diameters ranging from 0.082 to 0.375 inch.

Power for arc melting tungsten- and tantalum-base alloys in the furnace is provided by three 500-ampere ac arc welders operating on one phase of a three-phase 440-volt power line. Each welder is designed with power factor correction capacitors to operate at a power factor of approximately 75%. The welder consists essentially of a combination single-phase transformer and a movable core reactor—a transactor. The movable core is manually adjusted to control arc current during arc starting, ingot melting, and "hot topping" of a typical heat. Care is taken to adjust the cores of the transactors simultaneously so that power output is equalized and feedback avoided. The arc melting circuit consisted of the three transactors connected in parallel with the electrode and ground terminals of the furnace. A contactor is used in the secondary loop of each transactor so that they may be operated together or independently as power demand varies during the melting of a heat.

The pumping system used for evacuating the furnace consists of a 6-inch NRC B-6 booster diffusion pump and a Kinney KDH-130 mechanical forepump. The diffusion pump has

a pumping speed of 1000 cfm at 0.1 to 1.0 micron Hg, and the mechanical pump has a pumping speed of 130 cfm. Since the furnace volume is small (3.2 cu ft), the capacity of the pumping system was adequate for the intended melting program. Normal melting chamber pressure was 1 to 2 micron Hg. Normal leak rate was 10 to 12 microns/min. Pressure is read in the foreline and the vacuum line by gauges.

High-purity rods of tungsten, tantalum, molybdenum, and columbium were used as consumable electrodes for vacuum-arc melting of the alloys, and the chemical analyses and suppliers of these materials are given in Table II.

A novel multiple-electrode melting technique was employed for melting the selected refractory metal alloys. The number of rods in the electrode was determined by the number of component metals in the desired alloy; the diameter of each rod was determined by (1) the desired weight percentage of the component metal, and (2) the density of the component metal. To melt a given alloy of the W-Ta-Mo-Cb system, individual rods of appropriate diameters were pointed on one end to facilitate arc starting, and the opposite, unpointed ends of the rods were firmly clamped together in a tightly fitted cluster to form the consumable electrode.

The ingot molds were made of molybdenum and were lined with one layer of tungsten sheet (0.025 inch thick). The use of a high-melting mold material and the absence of water cooling resulted in consistently sound ingots.

After thorough chemical cleaning, the assembled electrode and the mold assemblies were placed in the furnace. The electrode was inserted into the electrically insulated electrode guide in the electrode housing which was tilted outward for electrode charging. The pointed end of the electrode was then lowered through the feed rolls and the electrical contact. Feed rolls were adjusted to firmly grip the largest of the electrode rods to feed the clustered rods during the arc melting sequence. After the electrode had been vertically aligned, the mold was placed on the hearth of the melting chamber so that it was concentric with the largest rod of the electrode. Since it was observed that the arc emanated from the largest rod during the melting of all of the alloys, the central location of this rod in the mold minimized arcing between the electrode and the mold. When

the electrode and the mold had been positioned and aligned, the furnace was closed and evacuated. The melting chamber pressure and leak rate of the system were determined. Power was turned on when the pressure reached 1 micron Hg.

Consumption of the multiple-rod electrode was started by gradually lowering the pointed end of the electrode to strike an arc with a deposit of tungsten or tantalum chips in the mold bottom. As the arc was started and the electrode was consumed, the lower melting point metals flowed to the tip of the rod with the largest diameter where a droplet formed. Even though the rods of the cluster might have become separated by as much as 3 or 4 millimeters, the droplet persisted as a result of strong surface tension forces. Constant agitation of the droplet was observed, and thus thorough mixing of the alloy components took place before the droplet reached a critical size and fell into the ingot pool.

As consumption of the electrode progressed, it was automatically fed downward. Electrode speed and arc length were controlled by arc voltage. When arc length increased, arc voltage correspondingly increased the electrode speed to shorten the arc length. The reverse of this sequence occurred when the electrode tip approached the ingot pool too closely. Arc power (for a given arc length) was adjusted by the furnace operator so that the ingot pool extended just to the mold wall. Excess power caused the molten metal to run through the mold or mold liner; insufficient power caused surface voids or cold shuts in the ingot. As the desired ingot length was reached, arc power was reduced so that the ingot pool was diminished gradually. In this way the liquid metal was allowed to freeze progressively from the bottom of the pool to the top and thereby eliminate solidification shrinkage cavities. Upon completion of melting, the ingot was slowly cooled in the furnace under vacuum.

When each ingot had cooled to room temperature, it was removed from the furnace, visually examined for defects, and photographed. Typical ingots of a tungsten-tantalum alloy produced for this project are shown in Figure 3.

Using the procedures and equipment described, ingots (2-1/2 to 3 inches long and 1-1/4 inches in diameter) of six alloys and unalloyed tungsten were successfully melted for Phase I.

W	Ta	Cb	Mo	Number Melted
25	75	--	--	6
--	88	--	12	6
44	44	12	--	8
88	--	6	6	8
68	20	--	12	6
20	68	--	12	6
100	--	--	--	1

Extrusion Experiments

The vacuum-arc-melted ingots were machined into extrusion billets by lathe turning and cylindrically grinding to a diameter of 1.000 ± 0.001 inch. Turned and ground ingots of the 68W-20Ta-12Mo and 68Ta-20W-12Mo alloys are shown in Figure 4. After the turning and grinding operations were completed, the top and bottom ends of the ingots were removed with abrasive cut-off wheels. To reduce the arc of contact between the abrasive wheel and the ingot, and thus minimize chances of fracture, the ingot was rotated as the cut-off wheel was fed into the material under a liberal flow of water. Two extrusion billets were cut from most ingots since previous experience had shown that better extrusions were obtained from billets with lengths ranging from 1 to 1-1/2 inches rather than from longer billets.

The top and bottom ends of the ingots that were cropped during the preparation of extrusion billets were prepared for hardness determinations and metallographic examination. Metallographic examination and hardness surveys (DPH) on the transverse sections did not show any evidence of macroscopic segregation or incomplete alloying in any of the ingots. The hardness values (average of two full-diameter traverses taken at 90 degrees to each other) for the selected alloys are given in Table III. The only hardness values that did not compare well were those for the 44W-44Ta-12Cb alloy; the hardness values for this alloy usually ranged from 393 to 421 DPH, whereas the values for Heats 226 and 227 ranged from 430 to 460 DPH.

Prior to extrusion, a slight chamfered nose was ground on each of the billets to facilitate entry into the die. Each billet was then grit-blasted and flame-sprayed with molybdenum to a thickness of approximately 15 mils to provide protection during heating and lubrication during extrusion.

Die inserts were forged from blanks of a hot-work steel (H13) , heat-treated to a hardness level of Rockwell "C" 52 to 54 and cylindrically ground. These die inserts were designed so that sheet-bar extrusions with round-cornered, rectangular cross sections (approximately 0.5 by 0.7 inch) could be obtained. To reduce die wash, the inner surfaces of the die inserts were grit-blasted, flame-sprayed with 5 mils of molybdenum, and then plasma-sprayed with 10 mils of zirconia. (Initial experiments had shown that the molybdenum-zirconia insert coating was superior to the previously used molybdenum-tungsten insert coating.) Typical sprayed extrusion billets and die inserts are shown in Figure 5.

The extrusion experiments were performed at Super Alloy Forge, Inc., Hamburg, Michigan, where a No. 1-1/2 National Maxipress (vertical press) was available. With the flywheel of the press operating at 100 rpm and a ram stroke of 6 inches, a free-running average ram speed of 1200 in./min was attainable. The press was capable of extruding billets up to 1 inch in diameter and 2 inches long through a die assembly placed on the lower platen.

The billets were placed on a tungsten stool and heated in a vertical induction coil that was cooled by high-pressure water and powered by a 9600-cycle, 100-kw generator. For extrusion, the unalloyed tungsten billets were heated to temperatures in the range of 3000 to 3200 F, the 88Ta-12Mo billets to 3200 to 3500 F, and the 75Ta-25W billets to about 3500 F. The 68W-20Ta-12Mo, 68Ta-20W-12Mo, 70Ta-19W-11Mo, 88W-6Mo-6Cb, and 44W-44Ta-12Cb alloys were heated to about 3800 to 4000 F. Since the billets were coated with molybdenum for protection during heating, heating was done in air. After the power was turned on, the total heating and soaking time was 2 to 3 minutes. The heated billets were transferred from the induction coil to the container, and the press was immediately actuated; the total elapsed time for this operation varied from 3 to 6 seconds.

Thirty-four billets were successfully extruded to sheet bars with round-cornered, rectangular cross sections (0.4 to 0.5 inch thick). Typical sheet-bar extrusions are shown in Figure 6. The results of the extrusion experiments given in Table IV may be summarized as follows:

Alloy	Number of Extrusions	Average Reduction of Cross-Sectional Area (%)	Average Yield (%)
25W-75Ta	9	62	81
88Ta-12Mo	10	61	80
44W-44Ta-12Cb	14	64	78
88W-6Mo-6Cb	10	62	76
68W-20Ta-12Mo	12	65	77
68Ta-20W-12Mo	12	66	84
70Ta-19W-11Mo	1	70	86
70Ta-19W-11Mo	1	76	86
22W-78Ta	4	61	80
100W	2	65	91

The average width of all extrusions (except 70Ta-19W-11Mo) was 0.70 inch. After the tails of the sheet bar extrusions were cropped and the noses ground to remove cracks or bursts, the billet-to-sheet bar yields were determined on the basis of weight. Most extrusion yields were quite good, except for a few instances when extrusion was incomplete because a follow block was not used. Since it was difficult to measure the area of a round-cornered, rectangular cross section, the approximate area was calculated with the following formula:

$$A_f = \frac{W_f l_o A_o}{W_o l_f}$$

where

A_f = cross-sectional area of the extrusion,
W_f = weight of the extrusion,

l_f = length of the extrusion,
l_o = length of the billet,
A_o = cross-sectional area of the billet,
W_o = weight of the billet.

Certain assumptions were made in these calculations and the volume of material removed in chamfering the nose of the billet was neglected; however, the approximate reduction of area figures are reasonable.

The average reduction of area value for all extrusions was 60 to 65% except for the 70Ta-19W-11Mo alloy. This off-composition alloy was melted for making tensile grip pins, and therefore was used to determine the feasibility of achieving larger reductions during extrusion. The extrusion trials showed that sheet bars with thicknesses of 0.38 and 0.30 inch (in contrast to the usual 0.44 inch) could be produced quite easily. Since the width of the sheet bars remained constant, the changes in reduction of area were not marked. Figure 7 shows an extrusion of the 68Ta-20W-12Mo (67% reduction of area) and two extrusions of the 70Ta-19W-11Mo (70 and 76% reduction of area).

Both the 68W-20Ta-12Mo and 68Ta-20W-12Mo alloys exhibited rather pronounced nose cracking. In future work, it would be desirable to use nose blocks in an attempt to eliminate severe nose cracking. The side wall tearing on some of the longer extrusions might be the result of an insufficient amount of molybdenum coating on the extrusion billet.

Representative as-extruded microstructures of the 75Ta-25W, 88Ta-12Mo, 68W-20Ta-12Mo, 68Ta-20W-12Mo, 44W-44Ta-12Cb, and 88W-6Mo-6Cb alloys are shown in Figures 8, 9, and 10.

Rolling Experiments

Experiments with Induction Heating

The results of extrusion experiments indicated that temperatures of approximately 2500 to 4000 F would be required for rolling the selected alloys. Provisions were

made for three methods of heating the samples for hot rolling. The first method consisted of placing the sample on a graphite or tungsten stool within a tightly wound, water-cooled induction heating coil that was insulated from the hot sample by a KT silicon carbide tube and alumina cement. This heating coil is very similar to that which had been used in the extrusion experiments where it was possible to attain temperatures in excess of 4000 F when 1-inch-diameter round extrusion billets were heated. However, it was foreseen that the sheet bar samples with smaller cross sections (0.5 by 0.7 inch) would be more difficult to heat and that the maximum attainable temperature would decrease as the cross-sectional area of the sample changed with each successive pass during rolling. It was also anticipated that supporting the sample in a vertical position might cause difficulties. Therefore a second induction heating coil with a tungsten susceptor was constructed for use in a horizontal position. The water-cooled coil was insulated from the tungsten susceptor with grain zirconia.

A third heating source—a molybdenum-wire-wound tube furnace—was constructed for heating samples in the temperature range of 2500 to 3000 F since the finishing temperatures for rolling the 88Ta-12Mo and the 75Ta-25W alloys would fall in that range. Two concentric alumina tubes were used in this furnace: the inner tube was wound with molybdenum wire and supported within the larger tube to allow for the flow of hydrogen for oxidation protection. The ends of the two tubes were sealed so that hydrogen flow was restricted to the space between the inner and outer tubes. With this arrangement, the sample could be heated in an inert atmosphere (such as argon) or in air but not in an atmosphere of hydrogen which would be detrimental to the alloys with high tantalum contents. As a safety precaution, the furnace was provided with a nitrogen flushing arrangement.

A small Stanat mill was used for the rolling experiments. A two-high roll configuration was used for this project. Six-inch-diameter hot-work rolls (H13) with 8-inch-wide faces were plasma-sprayed with about 10 mils of zirconia to provide a heat barrier between the steel roll surfaces and the hot sheet or sheet bar being rolled. Earlier rolling experiments at 2200 to 3000 F with unalloyed tungsten bars showed that even cold rolls were not subject to heat checking or softening when a zirconia coating was used. To

ensure good bonding of the zirconia, the rolls were first grit-blasted and sprayed with about 5 mils of molybdenum. The plasma-sprayed rolls are shown in Figure 11.

At the onset of the rolling experiments, numerous problems were encountered with the heating equipment. A burn-out was experienced in the molybdenum-wire-wound tube furnace. The furnace was later repaired and successfully brought up to 2800 F when the muffle tube cracked and thus leaked hydrogen. At this point, this method of heating was abandoned.

For the preliminary rolling experiments, the vertical induction coil was used for heating. The coil was powered by a 100-kw, 9600-cycle power source. The sheet-bar samples could be heated to 3500 F before the first pass, and the attainable temperature decreased as the thickness of the sample was reduced by rolling. For protection from oxidation, the sheet-bar samples (approximately 0.5 by 0.7 by 2 to 3 inches) were flame-sprayed with molybdenum, and, during heating, an argon flow was maintained inside the induction coil.

Attempts were made to hot roll sheet bars of the following alloys:

78Ta-22W*
88W-6Mo-6Cb
44W-44Ta-12Cb

The 78Ta-22W alloy was reduced 65% in thickness by rolling at temperatures in the range of 2700 to 3200 F. Although large reductions were possible at these temperatures, i.e., the material was inherently quite plastic, edge cracking was encountered. This edge cracking was attributed to contamination during heating and rolling. Rolling experiments on sheet bars of the 88W-6Mo-6Cb and the 44W-44Ta-12Cb alloys were unsuccessful; on the basis of the fracture characteristics, these failures were attributed to an insufficiently high heating temperature (3500 F). However, higher temperatures could not be attained with the vertical induction coil.

*An off-composition heat melted specifically for preliminary rolling trials.

Inability to attain sufficient temperatures for the rolling of the 88W-6Mo-6Cb and the 44W-44Ta-12Cb alloys with the vertical induction coil posed a major heating problem. Although the necessity of using 50-foot-long power leads from the power supply unit to work station was certainly not desirable, the large capacity (100 kw) of the unit was believed to be great enough to offset this disadvantage. Since flux linkage between the sample being heated and the induction coil appeared to be the limiting factor, an attempt was made to use the horizontal induction heating coil equipped with a tungsten susceptor. Efforts to properly balance the tank circuit to heat the susceptor were unsuccessful. Further work on this heating method was unnecessary because a plasma unit was installed in the Laboratory.

Experiments with Plasma Heating

Plasma heating was well suited to this application since the sample to be heated was relatively small. The temperatures required for rolling were relatively easy to attain because of the large amount of heat energy available from the dissociated and ionized plasma being emitted through the nozzle of the gun. A Metco Type 2M Plasma Flame Spray Gun with a Type B nozzle was used. Heating was done by passing the plasma back and forth over the sheet-bar sample which was placed on a graphite block in close proximity to the rolling mill. Nitrogen was used as the primary gas and hydrogen as the secondary gas. An illustration of the setup used for plasma heating and rolling is given in Figure 12.

Before proceeding with rolling experiments in which plasma heating was used, three protective coatings were obtained that were expected to be suitable for the 88Ta-12Mo and 75Ta-25W alloys. Since the compositions of these proprietary coatings cannot be revealed at this time, only the following designations will be used: X1, X2, and X3.

Coatings X1 and X2 were applied by dipping the conditioned, grit-blasted sheet-bar samples into a liquid dispersion of the coating material and subsequently drying in air. This step was repeated two to four times to obtain a coating weight of approximately 1/2 g/sq in.

After a sufficient amount of coating was applied, the sample was baked in vacuum at 1400 F for 30 minutes.

To obtain the triplex coating, X3, the first layer was applied to the sample in a manner similar to that used for X1 and X2 until a coating density of about 1/16 to 1/8 g/sq in. was achieved. The sample was then baked in vacuum at 1400 F for 30 minutes. Following the vacuum baking treatment, a coat of the second material was applied to the sample and allowed to dry in air; one coat of the third material was then applied to the sample and allowed to dry in air.

In the first group of rolling experiments in which plasma heating was used, three coated sheet-bar samples of each of the high-tantalum alloys (75Ta-25W and 88Ta-12Mo) were hot-rolled; for subsequent evaluation, a different protective coating was applied to each of the three sheet-bar samples of each alloy. Samples of the 75Ta-25W and 88Ta-12Mo alloys were reduced 52 to 80% in thickness in the temperature range of 3000 to 3600 F in 6 to 8 passes. Heating by plasma was very successful, and the protective coatings appeared to adhere quite well for at least 6 rolling passes. Throughout this investigation, the alloys were rolled directly from the as-extruded condition; no stress-relief or recrystallization annealing treatments were used.

Samples were taken from each of the six sheets for metallographic and hardness examination to determine which, if any, of the protective coatings were effective in preventing contamination. Microscopic examination did not reveal contamination in any of the samples, and numerous hardness probes (Knoop 100-gram load) showed that the maximum depth of contamination was approximately 0.005 inch. Coatings X1, X2, and X3 appeared to have performed equally well, but X1 was arbitrarily chosen for coating the remainder of the 75Ta-25W and 88Ta-12Mo alloys for rolling. A photomicrograph of a 75Ta-25W alloy that had been coated with X1 and then reduced 69% in thickness is shown in Figure 13. From this figure, it can be seen that there was virtually no difference in the size of the Knoop hardness indentations in material where the coating had adhered. Unfortunately, it is difficult to visually determine the degree of adherence of the coating to the sample after rolling since all of the coatings tend to spall upon cooling to room temperature.

On the basis of first trials with plasma heating, a schedule was devised for rolling the 75Ta-25W and 88Ta-12Mo alloys and unalloyed tungsten to intermediate gauge sheet 0.120 to 0.170 inch thick (Table V). The total number of passes was limited to 6 or 7 because on further rolling (1) the effectiveness of the protective coating was diminished and (2) plasma heating became more difficult as the sample became longer (limiting length for effective heating was about 5 inches).

Sheet-bar samples of 75Ta-25W, 88Ta-12Mo, and unalloyed tungsten were plasma-heated and successfully reduced to intermediate-gauge sheet except for one instance in which the roll setting was in error for the initial pass and a sample of 75Ta-25W was broken. In general, the final lengths of the sheets varied from 3-1/2 to 5 inches. These results showed that when protective coatings and plasma heating were used, very few problems were encountered in rolling the two high-tantalum alloys. The general appearance of the intermediate gauge sheets was satisfactory (Figure 14).

The intermediate-gauge sheet samples (nominally 0.7 inch wide and 0.1 to 0.2 inch thick) of 88Ta-12Mo, 75Ta-25W, and unalloyed tungsten that had been successfully rolled from sheet bars were conditioned for rolling to final gauge. All samples were grit-blasted, and edge cracks were removed by grinding; no material was removed from the rolling surface by grinding. In many instances, the intermediate-gauge sheet samples were cut in two to facilitate plasma heating during the further rolling operations. Coating X1 was applied to all samples since it had provided very good protection for the 88Ta-12Mo and 75Ta-25W alloys during the previous rolling experiments.

Before the final rolling operations on the 75Ta-25W and the 88Ta-12Mo alloys, it was necessary to establish the proper rolling temperature range for obtaining a warm-worked rather than a hot-worked (recrystallized) structure in these two alloys. To aid in the determination of the proper rolling temperature range, samples were taken from the intermediate-gauge sheets for metallographic examination in the as-rolled condition as well as in two annealed conditions. The annealing treatments were performed in vacuum at 2800 and 3000 F for 15 minutes.

A rough determination of the degree of recrystallization in each sample was made by microscopic examination. The results were as follows:

Alloy	Sheet Code	Degree of Recrystallization (%) As-Rolled	2800 F Anneal	3000 F Anneal
75Ta-25W	162T	70	90	100
	164T	60	75	100
	164B	25	35	50
	165	20	30	--
	182T	20	50	100
88Ta-12Mo	166T	20	30	75
	166B	30	70	100
	167B	30	40	75
	186T	20	30	100
	186B	20	40	100

The results of the microscopic studies showed that, in general, the reduction of the sheet bars to intermediate gauge sheets was accomplished by hot working. In addition, microscopic examination showed that recrystallization was 50 to 100% complete after 15 minutes at 3000 F. Since a warm-worked structure (no recrystallization) is desired in the final-gauge sheet material, the temperature range of 2500 to 3000 F was selected for rolling the intermediate-gauge sheets to final gauge.

After a few preliminary rolling trials, the rolling schedules shown in Table VI were chosen for rolling the 75Ta-25W and 88Ta-12Mo alloys and unalloyed tungsten to final-gauge sheet. Since the intermediate-gauge sheets were of various thicknesses, it was necessary to use different combinations of number of passes and rolling temperatures to achieve final sheet thicknesses of 0.060 to 0.080 inch. The lowest rolling (finishing) temperature for the 75Ta-25W and 88Ta-12Mo alloys was limited to 2600 F to avoid excessive edge cracking.

The rolling experiments were quite successful, and several intermediate-gauge sheets of the 75Ta-25W and 88Ta-12Mo alloys were reduced to final-gauge sheet (Figure 15). In addition, one sheet bar of 75Ta-25W (Code 183T) was rolled directly to final-gauge sheet and two intermediate-gauge sheets of unalloyed tungsten were rolled to final gauge. The performance of the protective coating (Coating X1) on the samples and of the zirconia-coated rolls was excellent. The results of these rolling experiments are given in Tables VII and VIII for 75Ta-25W and the 88Ta-12Mo alloy, respectively; these tables also indicate the reductions in thickness from sheet bar to intermediate gauge sheet and sheet bar to final gauge sheet.

Samples were taken from selected final-gauge sheets of the 75Ta-25W and the 88Ta-12Mo alloys for metallographic examination as well as chemical analyses. The results of chemical analyses showed that the levels of interstitial elements were tolerable, except for hydrogen. For example, the chemical analysis of one sample of the 75Ta-25W alloy was as follows:

Code	C	N	O	H
182T	21 ppm	5 ppm	13 ppm	8 ppm

However, a few experiments showed that the level of hydrogen in the 75Ta-25W or 88Ta-12Mo alloys could be easily reduced to about 2 or 3 ppm by vacuum outgassing for 1 hour at 1400 to 1800 F. Therefore, bend and tension test specimens prepared from these alloys were outgassed at 1600 F for 1 hour.

Attempts were made to roll three sheet-bar samples of each of the 88W-6Mo-6Cb and 44W-44Ta-12Cb alloys after plasma heating to approximately 4000 F. Only one rolling trial was even partially successful; a sheet-bar sample of 44W-44Ta-12Cb that had been coated with X1 was reduced 42% in thickness before it broke. Some other samples showed indications of being deformable by rolling even though they were broken on the first or second pass. The principal problem appeared to be that of finding an adequate protective coating. Not only does the sheer effect of temperature have to be considered, but also the erosive effects of the plasma in heating to attain and hold the sample at 4000 F.

Sacrificial coatings of molybdenum and a glass-type coating were grossly ineffective. Coating X1 was effective for only the one sample cited in the foregoing.

Oxidation of the subject alloys is catastrophic at temperatures above 3500 F when the protective coating breaks down. The microstructure of a sample of the 44W-44Ta-12Cb alloy after heating to approximately 3800 to 4000 F and rolling to a thickness reduction of 42% (Figure 16) shows severe grain-boundary oxidation and cracking.

An evaluation of various protective coatings was undertaken before any further rolling was done on the 44W-44Ta-12Cb, 88W-6Mo-6Cb, 68W-20Ta-12Mo, and 68Ta-20W-12Mo alloys. For this evaluation, each coating was applied to a sample of wrought tungsten (shape similar to sheet bar) and subjected to plasma heating.

Since tungsten oxide is volatile at temperatures above about 2600 F, breakdown of the protective coating would be detected by evolution of white smoke from the sample. The edges and corners of the tungsten samples were rounded by grinding and were subsequently grit-blasted and degreased with Chlorothene so that maximum coating adherence could be obtained.

Twelve protective coating systems were evaluated. Since these coatings are proprietary, only the following designations will be used:

X1	ZrO_2 + X3
X2	ZrO_2 + X6
Al_2O_3 + X3	ZrO_2 + X7
Al_2O_3 + X6	$MoSi_2$-ZrB_2 + X6
Al_2O_3 + X7	$MoSi_2$-ZrB_2 + X7
Al_2O_3 + ZrO_2 + X3	$MoSi_2$-ZrB_2 + X3

The Al_2O_3, ZrO_2, and $MoSi_2$-ZrB_2 were applied to the tungsten samples by plasma spraying to thicknesses of 5 to 10 mils. With the exception of X6, the coatings designated X- were applied by dipping the samples into a liquid dispersion of the coating material and subsequently drying in air. This step was repeated 2 to 4 times to obtain a coating of adequate thickness. After a sufficient amount of coating had been applied, the samples were baked in

in vacuum at 1200 F for 30 minutes to minimize oxygen contamination and to yield maximum density and adhesion for protection against the high velocity of the plasma flame used for heating the samples. Coating X6 was heated until molten and the sample was coated by dipping; no vacuum treatment was required.

None of the coatings was protective at temperatures above about 3500 to 3600 F. The criterion for coating breakdown, namely, evolution of tungsten oxide, made evaluation quite simple. For the coatings that contained $MoSi_2$, it was difficult to determine whether white smoke was being evolved from the tungsten sample, the molybdenum-containing coating, or both. When this coating was applied to a tantalum sample, smoke was evolved at the same temperature as for the tungsten sample (indicative of the breakdown of $MoSi_2$); therefore, it was decided that no further work would be done with coatings that contained $MoSi_2$.

Seven additional coatings were evaluated:

X3 + X4 + X5
X1 + X5
X1 + X9
X10
X11
Al_2O_3 + ZrO_2 + X8
Al_2O_3 + ZrO_2 + SiO_2 + X8

From this group, X11 was selected for coating the samples for the final rolling experiments since it appeared to perform better than the other coatings.

Sheet-bar samples of each of the following alloys were coated with X11:

88W-6Mo-6Cb
68W-20Ta-12Mo
68Ta-20W-12Mo
44W-44Ta-12Cb

The coating procedure was the same as that described for X1. These samples were plasma heated to temperatures in the range of 3500 to 4000 F for rolling, but all of them failed. Reductions of thickness as high as 20% were achieved before the samples failed by cracking, and metallographic examination

revealed that the temperatures were sufficiently high to promote recrystallization during rolling. It is felt that better protective coatings will have to be developed before these alloys may be successfully rolled when the plasma heating technique is used. However, other combinations of heating and processing techniques may be satisfactory for fabrication of these alloys.

A summary of the melting, extrusion, and rolling for Phase I is given in Table IX.

Metallographic Preparation

The metallographic preparation of tungsten- and tantalum-base alloys presented difficulties similar to those experienced with Group IVa and other Group Va and VIa metals. Since these metals and their alloys have a tendency to flow, care was required during sectioning, grinding, and polishing in order that the flowed metal developed in each step could be readily removed in the following step. With the tantalum-base alloys in particular, there was also a tendency for grinding scratches to widen (open up) during the final polishing operation with the resultant formation of pits. Proper technique involves reducing polishing time to a minimum by keeping the sample size reasonably small (1/16 inch square is satisfactory) and by carrying grinding through 4/0 emery paper.

The following metallographic preparation sequence was employed in this program:

(1) Sectioning and Mounting.—A water-cooled Allison WA90KRA cut-off wheel permitted fast cutting without overheating. Specimens were molded in Bakelite epoxy resin which withstood attack by the mixed-acid etchant that was used.

(2) Grinding.—For rough grinding, a 120-grit SiC wet belt sander was used followed by 360, 400, 500, and 600-grit SiC abrasive papers. Fine grinding was done on 3/0 and 4/0 emery papers.

(3) Polishing.—Hand polishing was employed with Linde A abrasive on Forstmann cloth. Moderately heavy pressure with low speed gave best results (the Automet polisher also gives satisfactory results).

(4) Etching.—The following mixed-acid etchant was used with tantalum-base alloys and with tungsten-base alloys containing more than a 10% total of Ta, Cb, and Mo:

Lactic Acid (85%)	45 cc
HCl (38%)	35 cc
HNO_3 (71%)	30 cc
HF (49%)	10 cc

Etching action is slow, requiring 1 to 3 minutes. When etching revealed disturbed metal (usually in the form of stained unresolved structure), the final polishing and etching steps were repeated. The etchant above ages rapidly and for this reason it should be discarded 1/2 hour after mixing. If the etchant is not fresh, there is a loss of grain boundary contrast in the etched specimen.

The following etchant was used for unalloyed tungsten:

$CuSO_4$	10 g
H_2O	40 cc
NH_4OH (conc.)	20 cc

Application was by swabbing for about 1 minute.

Recrystallization Studies

Of the six alloys selected for Phase I, only the 75Ta-25W and 88Ta-12Mo alloys were successfully rolled to final-gauge sheet. Also included in the rolling was a high-purity tungsten heat for control purposes. The reduction of sheet bars to intermediate gauge sheets was, in general, accomplished by true hot working, but the reduction of intermediate-gauge sheet samples to final gauge was achieved by rolling at temperatures where warm worked (unrecrystallized) structures were obtained. Representative microstructures of intermediate- and final-gauge sheet samples of the alloys are shown in Figures 17 and 18.

Before specimens were prepared for recrystallization studies and mechanical tests, final-gauge sheet samples were subjected to hardness and metallographic examination as well as chemical analysis to determine whether the selected as-ground sheet thickness of 0.050 inch would be acceptable from the standpoint of contamination. The chemical composition of a representative sheet (184-1) of the 88Ta-12Mo alloy before and after grinding was as follows:

Condition	C (ppm)	O (ppm)	N (ppm)	H (ppm)	Mo (wt %)
As-rolled (0.070 inch thick) and grit-blasted	26	130	47	10	11.2
Ground to 0.050 inch thick	6	26	42	9	--

The carbon, oxygen, and nitrogen contents were reasonably low, but the hydrogen content was deemed to be too high. However, as shown previously for the 75Ta-25W alloy, vacuum degassing for 1 hour at 1400 and 1800 F reduced the hydrogen contents of 0.050-inch-thick sheet samples to 3 and 2 ppm, respectively. Based on these results, 0.050 inch was selected as the thickness of all specimens to be subjected to recrystallization studies and mechanical tests.

For recrystallization studies, 0.050-inch-thick specimens were cut from two sheet samples (184-2 and 186T) of the 88Ta-12Mo alloy, from three sheet samples (160-1, 162B, and 183T) of the 75Ta-25W alloy, and from sheet samples of unalloyed tungsten. Vacuum annealing treatments were performed in a resistance-heated tantalum tube furnace enclosed in a bell jar. All tantalum alloy specimens were heated to 1600 F, held for 1 hour for outgassing, and then heated to the required annealing temperature and held for 1 hour. The outgassing stage was eliminated in tungsten specimens as unnecessary. Vacuum chamber pressures during heating and holding at temperature ranged from 5×10^{-4} to 1×10^{-5} mm Hg.

The amount of recrystallization in each sample was determined metallographically at 100X on the longitudinal section (i.e., in the rolling direction). In addition, DPH readings were taken on a Tukon tester. The results of the recrystallization studies are given in Tables X, XI, and XII and Figures 19, 20, and 21. The approximate recrystallization temperatures (temperature at which the structure was 50% recrystallized) for the 75Ta-25W, 88Ta-12Mo, and unalloyed tungsten sheet were 2850, 2600, and 2425 F, respectively. The variation in recrystallization behavior from heat to heat was small; however, there were a few inconsistencies in the hardness data. Based on the recrystallization data, 2300 F was selected as the stress-relieving temperature for mechanical test specimens of the 75Ta-25W and 88Ta-12Mo alloys and 1900 F as the stress-relieving temperature for the unalloyed tungsten.

The recrystallization temperature of 2425 F for tungsten sheet indicates a moderately heavy degree of warm work. For comparison, recrystallization values ranging from 2425 to 2575 F, corresponding to various degrees of warm working, were reported by Crimmins[2] on powder-metallurgy tungsten sheet. Additions of tungsten or molybdenum to tantalum raised the 50% recrystallization temperature of tantalum well above that of the unalloyed components (Mo, W, and Ta). This feature is summarized in Table XIII. An examination of these data also reveals molybdenum to be more potent than tungsten in raising the recrystallization temperature of tantalum.

Bend Transition Studies

Bend transition temperatures were determined for all alloys that had been successfully warm-rolled to final-gauge sheet: unalloyed tungsten, 75Ta-25W, and 88Ta-12Mo. Bend specimens were cut longitudinally and prepared by first grinding to size (0.6 by 1.2 by 0.045 to 0.055 inch) and then surface finishing to 400-grit SiC. Typical 75Ta-25W and 88Ta-12Mo bend specimens are shown in Figure 22. All specimens were stress-relieved in vacuum for 1 hour prior to testing. Stress-relief temperatures were 2300 F for the 75Ta-25W and 88Ta-12Mo alloys and 1900 F for unalloyed tungsten.

---

[2] P. P. Crimmins and C. W. Heimlich, "Spinning of Refractory Alloys," Metal Progr. pp 67-72 (December 1962).

The arrangement that was used for bend testing is illustrated in Figure 23. An interchangeable punch of the required 4T* radius was secured to the crosshead of a universal testing machine, and an interchangeable die, of required 12T span and 1.5T radius, was positioned axially with the punch. A split resistance furnace enclosed the assembly and was controlled to ±10 deg F throughout the test. Specimens were held for 15 minutes at temperature prior to testing. The loading rate was held constant at a crosshead travel speed of 0.025 in./min.

The ductile-brittle transition temperature in Group VIa metals has been shown to be related to metal purity and to the degree of cold work[3-8] with high purity and cold work operating to extend low-temperature ductility. The bend transition temperature of 450 F obtained on tungsten sheet (Figure 24) compares favorably with a value of 600 F obtained by Crimmins[2] on cold worked powder-metallurgy sheet and by Harmon[8] on arc-melted, hot-swaged rod. The lower value of 450 F is presumably attributable to higher purity and/or greater degree of cold work.

The high transition temperatures of the tantalum alloys (325 F for 88Ta-12Mo, Figure 25; and 450 F for 75Ta-25W, Figure 26) were disappointing. Schmidt[9] reported excellent bend ductility (0T bends) at both room temperature and at -320 F for unalloyed tantalum and for 90Ta-10W alloy. The

*4T = 4 x thickness of the specimen.

[3]D. McLean, Grain Boundaries in Metals, Oxford University Press, 1957.

[4]J. H. Bechtold, "Strain Rate Effects in Tungsten," J. Metals 8, 142-146 (1956).

[5]J. H. Bechtold, "Tensile Properties of Annealed Tantalum at Low Temperatures," Acta Met. 3, 249-254 (1955).

[6]R. H. Atkinson (Office of Ordnance Research, U. S. Army), "Tungsten Alloy Development," paper delivered at Conference on Government-Sponsored Research in Progress on Tungsten, Durham, North Carolina (May 20, 21, 1959).

[7]J. H. Bechtold, E. T. Wessel, and L. L. France, "Mechanical Behavior of the Refractory Metals," in Refractory Metals and Alloys, Interscience Publishers, Inc., New York, 1961, pp. 25-82.

[8]E. L. Harmon, "Investigation of the Properties of Tungsten and Its Alloys," WADD TR 60-144 (May 1960).

[9]F. F. Schmidt et al., "Investigation of the Properties of Tantalum and Its Alloys," WADD TR 59-13 (March 1960).

sharp rise in transition temperature between 90Ta-10W and 75Ta-25W was unexpected but was apparently the result of a sharp drop in the tolerance of tantalum for interstitials within the 10 to 25% tungsten range. Molybdenum was even more potent than tungsten in raising the ductile-brittle transition temperature of tantalum. Accordingly, the addition of 12% molybdenum raises the transition temperature of tantalum from -320 F to 325 F. These results are surprising considering that molybdenum has a somewhat greater interstitial solubility and lower transition temperature than does tungsten. Bend-transition data are plotted together in Figure 27 and given in Table XIV to facilitate comparison.

Future work toward lowering the ductile-brittle transition temperature of refractory alloys should include consideration of addition of rhenium[10]; addition of scavenger elements, such as zirconium and aluminum[11]; and addition of grain-refining dispersed oxides,[12] such as zirconia and thoria.

Tension Tests

Elevated-temperature tension tests were performed in vacuum on specimens prepared from final-gauge sheet samples of 75Ta-25W, 88Ta-12Mo, and unalloyed tungsten. The tension test specimens were cut longitudinally and prepared by grinding to size (0.600-inch-long, 0.150-inch-wide, 0.030 to 0.050-inch-thick gauge section) and then surface finishing to 400-grit SiC. Typical tensile specimens are shown in Figure 28. All specimens were stress-relieved in vacuum for 1 hour prior to testing. The stress-relief temperatures were 2300 F for the 75Ta-25W and 88Ta-12Mo alloys and 1900 F for unalloyed tungsten.

---

[10] R. I. Jaffee, D. J. Maykuth, and R. W. Douglass, "Rhenium and the Refractory Platinum-Group Metals," in Refractory Metals and Alloys, Interscience Publishers, Inc., New York, pp. 383-463.

[11] E. P. Abrahamson, II, and N. J. Grant, "Chromium Base Alloys," Parts I, II, and III, U. S. Navy Bureau of Aeronautics, Report NOas 56-1090-d (1957).

[12] J. L. Ratliff, D. J. Maykuth, H. R. Ogden, and R. I. Jaffee, "Development of a Ductile Tungsten Sheet Alloy," U. S. Navy Bureau of Weapons, Summary Report NOw 61-0677-c (May 1962).

For testing, a vacuum furnace with resistance-heated tantalum elements and an auxiliary vacuum pumping apparatus and power supply were adapted to a Riehle FS-30 Screw Powered Universal Testing Machine. The load was applied through pull rods that were attached to spherical seats in the top and bottom crossheads of the testing machine. The pull rods are introduced into the vacuum chamber through water-cooled "O" ring vacuum seals.

Each specimen was thoroughly cleaned and placed in the grip assembly (Mo-0.5Ti grips and tungsten adapters) within the vacuum chamber which was evacuated to a pressure of 1×10^{-5} mm Hg. The cold leak rate was approximately 0.5 to 1 micron/hr. Any tare load registered on the machine was removed by balancing the load indicator. Tare loads due to friction between the pull rods and "O" ring vacuum seals during testing were within the accuracy of the testing machine (1% of the load range used).

Because of the short duration of the test, the furnace power was controlled manually. Each specimen was brought to temperature within 40 minutes and held for 15 minutes before the test was started. The temperature of the specimen was measured with either two W—W, 26% Re or two Pt—Pt, 10% Rh thermocouples. The welded hot junction of each thermocouple was tied to either end of the 1/2-inch gauge length of the specimen. The pressure was not allowed to exceed 7×10^{-4} mm Hg during the heating time. A testing machine crosshead travel speed of 0.02 to 0.03 in./min was used throughout each test.

The results of the tension tests are given in Table XV. The 3000 F tensile strengths of the 75Ta-25W and 88Ta-12Mo alloys and unalloyed tungsten agree quite well with previous results on as-extruded bar materials (Table I). The tensile strengths of the 75Ta-25W and 88Ta-12Mo alloys markedly decreased from 3000 to 3500 F; however, the 75Ta-25W alloy still had a strength advantage over unalloyed tungsten at 3500 F whereas the tensile strength of the 88Ta-12Mo alloy was nearly the same as that of unalloyed tungsten.

Summary

Work under a previous contract resulted in the development and evaluation of several alloys of the W-Ta-Mo-Cb system, and many of these alloys exhibited extraordinarily high strength properties at 3000 F. The objective of the first

phase of the present contract was to produce, process to experimental sheet form, and evaluate at least six of these alloys. Strength and fabricability were the basis upon which the selections were made from the twenty W-Ta-Mo-Cb alloys studied under Contract AF33(616)-6172. Because previous extrusion experience had indicated that increased high-temperature strength made workability more difficult, both high-strength and intermediate strength alloys (respectively, >50,000 psi and 25,000 to 50,000 psi tensile strength at 3000 F) were chosen:

88Ta-12Mo	88W-6Mo-6Cb
75Ta-25W	68W-20Ta-12Mo
68Ta-20W-12Mo	44W-44Ta-12Cb

Small cylindrical ingots (1-1/4-inch diameter) of each of the alloys were consumably arc melted (ac power) by the multiple electrode technique. One-inch-diameter billets were then prepared from the ingots and all of the billets were induction heated and impact-extruded into sheet bars with round-cornered, rectangular cross sections (approximately 0.5 by 0.7 inch). The average reduction of area for all extrusions was 60 to 65%. The extrusions were made at temperatures of 3000 to 4000 F.

For hot rolling the sheet-bar samples to sheet form, provisions were made for three methods of heating: a vertical induction coil, a horizontal induction coil with a susceptor, and a molybdenum-wire-wound tube furnace. When efforts to employ these heating devices were unsuccessful, plasma heating was adopted. Plasma heating was well suited tothis application since the sample to be heated was relatively small. Rolling was accomplished with six-inch-diameter hot-work rolls which were plasma-sprayed with zirconia to provide a heat barrier between the roll surfaces and the hot sheet or sheet bar being rolled.

Several protective coatings were evaluated for application to sheet bars. Suitable coatings were found for 75Ta-25W and 88Ta-12Mo, and these alloys were successfully rolled to sheet. But, none of the protective coatings was satisfactory at temperatures above 3500 F— temperatures that were required for the breakdown of the other four alloys. Other combinations of heating and processing techniques may be satisfactory for fabrication of these alloys; however, further coating development is necessary before plasma heating can be utilized.

Recrystallization temperatures were determined for the 75Ta-25W and 88Ta-12Mo alloys as well as for unalloyed tungsten. The values obtained (2850 F for 75Ta-25W, 2600 F for 88Ta-12Mo, and 2425 F for unalloyed tungsten) indicated that these materials had received an appreciable amount of warm working. Molybdenum appeared to be more potent than tungsten in raising the recrystallization temperature of tantalum.

When sheet specimens were tested in vacuum at 3000 F, the tensile strengths of the 75Ta-25W and 88Ta-12Mo alloys and unalloyed tungsten compared favorably with previously determined values for as-extruded materials. The strengths of both alloys decreased markedly from 3000 to 3500 F. However, the 75Ta-25W alloy still had a strength advantage over unalloyed tungsten at 3500 F whereas the tensile strength of the 88Ta-12Mo alloy was nearly the same as that of unalloyed tungsten.

Bend test results showed that the transition temperatures for the two tantalum-base alloys were relatively high and tend to indicate that the tolerance of tantalum for interstitial elements is appreciably reduced by the presence of molybdenum and tungsten.

PHASE II

Melting Experiments

In Phase II of this investigation, at least one composition within the quaternary system W-Ta-Mo-Cb was to be further alloyed with additions that might strengthen the base composition by a dispersion effect. The reactive elements titanium, zirconium, and vanadium in combination with carbon were considered to be suitable and were therefore studied.

In the initial melting experiments under Phase II, techniques were studied for the addition of very small amounts of reactive elements and carbon to the melt. The three possible melting methods for the preparation of the Phase II alloys were as follows:

(1) nonconsumable arc melting;

(2) consumable vacuum-arc melting of a pressed and sintered electrode;

(3) consumable vacuum-arc melting by the multiple electrode technique.

Past experience at Crucible had indicated that nonconsumable arc melting produces unsatisfactory soundness, purity, and workability in the resulting ingots. On the other hand, the use of pressed and sintered electrodes was not compatible with our existing melting equipment. Therefore, consumable vacuum-arc melting by the multiple electrode technique was adopted for the preparation of these alloys.

In the first experiment, the feasibility of adding titanium to tungsten by melting a tungsten electrode into a mold lined with titanium sheet was studied; however, this procedure was discarded because premature melting of the titanium liner led to gross ingot inhomogeneity.

The most encouraging technique for alloy addition consisted of simultaneously feeding two electrodes: one electrode was pure tungsten (0.250-inch-diameter) and the other electrode was composed of four zirconium wires (0.010-inch-diameter) and tungsten carbide powder contained in a titanium tube (0.125-inch OD by 0.100-inch ID). A

1-1/4-inch-diameter by 3-inch-long ingot of a W-Ti-Zr-C alloy was obtained by consumable vacuum arc melting the multiple electrode into a split molybdenum mold lined with 0.020-inch-thick tungsten sheet; ac power was used. It was noted that the melting characteristics changed markedly from the bottom to the top of the ingot.

After the tungsten sheet mold liner was removed, the ingot was sectioned for metallographic examination and hardness determinations. Metallographic examination showed that complete melting was achieved; however, a high degree of porosity was noted in the bottom portion of the ingot. No gross segregation or unmelted tungsten carbide particles were visible; the microstructure consisted of grain boundary carbides and large columnar grains. Hardness measurements were quite uniform across the ingot cross section.

A portion of the W-Ti-Zr-C alloy ingot was analyzed for chemical composition. The approximate chemical composition of the multiple electrode materials and the results of the chemical analysis of the ingot were as follows:

	C	Ti	Zr	O	H	N	W
Composite Electrode	0.38	1.93	0.19	-	-	-	bal.
Ingot (Heat 190)	0.17	0.08	0.09	15 ppm	2 ppm	1 ppm	bal.

Slightly less than one-half of the carbon and zirconium were recovered in the ingot and only 4% of the titanium was recovered. The high titanium loss probably accounts for the porosity in the ingot. The oxygen, nitrogen, and hydrogen contents were maintained at acceptable levels.

Because of poor titanium recovery when a titanium tube was employed, titanium carbide was used in the next attempts to melt a W-Ti-Zr-C alloy. It was expected that the titanium recovery would improve since the carbide has a higher melting point than the pure metal. In the two melts made with titanium carbide, the bottom portions of the ingots were porous, and the molds were not completely filled near the bottom. However, the top portion of each mold was completely filled, and the top sections of the resulting ingots were dense. The homogeneity of the top portion of one of the ingots is shown by the micrographs in Figure 29. Increased fluidity and lower resultant porosity in the later stages of the melts were attributed to a marked increase in mold temperature as melting progressed.

A portion of one of the W-Ti-Zr-C alloy ingots was analyzed for chemical composition. The calculated chemical composition of the electrode materials and the results of the chemical analyses of the ingot were as follows:

	C	Ti	Zr	W
Electrode Material	0.38	3.56	0.20	bal.
Ingot (Heat 205)	0.40	0.08	0.16	bal.

The titanium recovery in the ingot was still very low (about 2%) even though titanium carbide (rather than elemental titanium as in the previous trials) was used in melting.

At this point in the program the base composition of the Phase II alloy was chosen before further melting experiments were undertaken. An alloy base of 88W-12Cb was selected; further alloying additions to be studied were to include small amounts of titanium, vanadium, zirconium, and carbon. The compositional aim was to be W-12Cb-1.0Ti-0.1Zr-0.05C; however, since the recovery of titanium in melting alloys of this type had been very low, consideration was also to be given to preparation of a titanium-free alloy or replacement of the titanium with vanadium.

Little difficulty was encountered in the consumable-electrode vacuum-arc melting of two ingots of a tungsten alloy with nominally 12% columbium, 0.1% zirconium, and 0.1% carbon. The electrodes were a solid tungsten rod and a columbium tube filled with zirconium wires and tungsten carbide powder. The melting characteristics of this alloy were very similar to those of an alloy melted from solid tungsten and solid columbium electrode rods. The columbium, and presumably the zirconium, melted 1 to 2 inches above the tip of the tungsten electrode and flowed to the tip of the electrode before dropping into the melt. Solid particles, undoubtedly tungsten carbide, could be observed in the liquid metal coating the tungsten electrode. Even though solid tungsten carbide particles were observed on the electrode, metallographic examination showed that complete melting was achieved in the ingot (Figure 30).

The two W-12Cb-0.1Zr-0.1C ingots were analyzed for chemical composition. The calculated electrode composition and the results of chemical analyses of the ingots were as follows:

	C	Cb	Zr	W
Electrode material	0.36	11.30	0.16	bal.
Ingot (Heat 206)	0.23	9.94	0.14	bal.
Ingot (Heat 207)	0.19	10.68	0.14	bal.

In a further effort to retain titanium in the ingot, a nominal W-12Cb-1.0Ti-0.10Zr-0.10C alloy was melted. The composite electrode was a columbium tube that contained titanium carbide powder and zirconium wires. A tungsten rod was used as the other electrode. The chemical analysis of the resulting ingot and the calculated electrode analysis were as follows:

	C	Cb	Ti	Zr	W
Electrode material	0.16	11.75	0.70	0.18	bal.
Ingot (Heat 209)	0.13	10.92	0.09	0.14	bal.

Inasmuch as titanium recovery in all melting trials—including the tungsten as well as the W-12Cb alloy—was very low, an attempt was made to substitute another strong carbide former, vanadium, for titanium. An ingot was made by simultaneously melting a tungsten electrode and a composite electrode (columbium tube, vanadium wire, zirconium wire, and tungsten carbide powder) of nominal composition W-12Cb-1.6V-0.13Zr-0.10C. The ingot analysis and calculated electrode composition given below indicated an appreciable vanadium recovery:

	C	Cb	V	Zr	W
Electrode material	0.075	11.84	1.61	0.13	bal.
Ingot (Heat 229)	0.07	12.59	0.29	0.12	bal.

On the basis of this vanadium recovery value, another ingot was melted to give a final vanadium content of 1.0%. The calculated electrode composition and the ingot analysis were as follows:

	C	Cb	V	Zr	W
Electrode material	0.072	11.20	6.09	0.12	bal.
Ingot (Heat 233)	-	-	1.21	-	bal.

Since 88W-12Cb was the base composition for the Phase II alloy and wrought 88W-12Cb material would be required for determination of base line data, three ingots of this alloy were also consumable-electrode vacuum-arc melted.

Vacuum-arc melting of consumable multiple electrodes has proved to be an effective method for adding small amounts of reactive metals and carbon to refractory metal alloys. However, the carbide-forming elements (titanium and vanadium) cause difficulty in that their vapor pressures, at temperatures encountered during melting, are extremely high. Poor recovery and porosity evidently are serious problems with titanium, but the substitution of vanadium has resulted in higher recovery and an apparent reduction in porosity.

Extrusion Experiments

Extrusion billets were prepared from the Phase II alloy ingots and three ingots of the 88W-12Cb alloy that had been melted to provide base line data. Billet preparation was identical with that described previously in this report. As in Phase I, the extrusion experiments were performed at Super Alloy Forge; however, round-bar rather than sheet-bar extrusions were produced. The die inserts were machined with a 70-degree included entrance angle and an inside diameter of 0.56 inch so that a reduction ratio of approximately 3 to 1 could be obtained. All billets were heated to approximately 4000 F for extrusion.

The results of the extrusion experiments are given in Table XVI. Except for Heat 219T, the 88W-12Cb alloy extruded easily, the extrusion yields were high, and the resultant extrusions were sound with good side walls (Figure 31). The extrusion experiments with the W-12Cb-Zr-C (Heat 207) and W-12Cb-V-Zr-C (Heat 229) were very successful; except for the side wall tearing of Heat 229, the extrusions were sound (Figure 32). Due to poor as-cast structure, the second W-12Cb-V-Zr-C billet (Heat 233) was not extruded satisfactorily.

Metallographic Studies

Metallographic studies were conducted on selected Phase II alloys for the purpose of comparing recrystallization behavior as well as carbide solubility. Both as-extruded and solution-annealed (3500 F for 1 hour, tin bath quenched) samples were examined.

A bell-jar vacuum furnace, operated at about 10^{-4} mm Hg, was used for the solution-annealing treatments. The arrangement, illustrated in Figure 33, permitted specimens to be suspended in the furnace by means of a tungsten wire. The end of the wire was attached to a piece of ferritic stainless steel that was in turn held by an electromagnet. The specimen was heated by radiation from a cylindrical tantalum resistance element positioned concentrically inside the tantalum and stainless steel radiation shields. Upon completion of the soaking period, the electromagnet was de-energized, and the sample fell freely into a molten tin bath maintained at 700 F.

Micrographs (100 and 1000X) of the 88W-12Cb, W-12Cb-Zr-C, and W-12Cb-V-Zr-C alloys in the as-cast, as-extruded, and solution-annealed (3500 F) conditions are shown in Figures 34, 35, 36, and 37. Comparison of the as-cast and the as-extruded microstructures reveals that the extrusion operation effected a good breakdown of ingot structure. All three alloys were substantially warm-worked, and only a small amount of recrystallization was observed in the W-12Cb-V-Zr-C alloy (Figure 37). The 1000X micrographs are of particular interest because they reveal the morphology of the carbides. As expected, the 88W-12Cb base line alloy (Figure 35) was free from carbides; however, the Zr-C and V-Zr-C alloys contained carbides that differed in size and shape. The carbides in the W-10.7Cb-0.14Zr-0.19C alloy (Figure 36) were massive and lamellar, whereas the carbides in the W-12.6Cb-0.29V-0.12Zr-0.07C alloy (Figure 37) appeared as dark clouds of fine discrete particles.

The microstructures of the samples that had been solution-annealed at 3500 F for 1 hour and quenched in a tin bath revealed important recrystallization and carbide solubility features. The solution-annealed 88W-12Cb alloy was fully recrystallized, whereas the solution-annealed Zr-C-containing and V-Zr-C-containing alloys were both less than 50% recrystallized. Significantly, annealing at 3500 F effected only minor solutioning of the massive carbides in the W-10.7Cb-0.14Zr-0.19C alloy, but there appeared to be complete solution of the fine carbides in the lower carbon alloy, W-12.6Cb-0.29V-0.12Zr-0.07C. Thus, in this series of alloys, carbon at the 0.07% level raised the recrystallization temperature above 3500 F; also, some degree of carbide supersaturation resulted upon quenching from 3500 F. Increasing the carbon content to 0.19% provided no additional increase in the recrystallization temperature, and the massive carbides were not taken into solution at 3500 F. It is realized that the addition of vanadium may have an effect on the solubility of carbon in the 0.19C alloy. Since the role of vanadium is not understood, the study of the effects of vanadium level on carbide solubility and strength properties would provide an interesting area for further research.

Mechanical Tests

One shouldered-end, button-head tension test specimen (Figure 38) was prepared from each of the following extrusions for testing in vacuum at 3500 F:

Heat 218B	88W-12Cb
Heat 207T	W-12Cb-Zr-C
Heat 229	W-12Cb-V-Zr-C

The testing equipment was described in Phase I.

Because of the short duration of the test, the furnace power was controlled manually. Each specimen was brought to temperature within 40 minutes and held for 15 minutes before the test was started. The temperature of the specimen was measured with W—W,26% Re thermocouples. The pressure was not allowed to exceed 7 x 10^{-4} mm Hg during the heating time. A testing machine crosshead travel speed of 0.02 to 0.03 in./min was used throughout each test.

The results of the tension tests on the as-extruded materials at 3500 F in vacuum are given in Table XVII. To our knowledge, the tensile strength of 27,000 psi for the base composition of 88W-12Cb (Heat 218B) is higher than any 3500 F tensile strength value reported to date. However, the W-12Cb-Zr-C alloy exhibited a tensile strength of 49,000 psi, which indicates that small additions of zirconium and carbon to the 88W-12Cb base composition result in very significant strength increases. In fact, the 3500 F tensile strength of the W-12Cb-Zr-C alloy is nearly identical with the tensile strength of the 88W-12Cb alloy at 3000 F. Initially, the tensile strength of the W-12Cb-V-Zr-C alloy (Heat 229) could not be determined due to failure of the grips; when the test was discontinued, the load corresponded to a stress of 55,000 psi on the specimen. When the vacuum chamber was opened, it was observed that the grips had undergone extensive deformation; however, the specimen was not fractured and had not deformed a measurable amount. After new tungsten grips had been prepared, the W-12Cb-V-Zr-C alloy specimen was retested at 3500 F, and the tensile strength obtained (57,000 psi) was slightly over twice that of the 88W-12Cb base composition. Prior stressing may have had an effect on the tensile strength of the W-12Cb-V-Zr-C alloy; however, if so, it would have only been deleterious.

Even more interesting than their effect on tensile strength was the effect of the small amounts of reactive elements and carbon on the ductility of the 88W-12Cb composition at 3500 F. The elongation and reduction of area of the base composition at 3500 F were less than 1%; moreover, at 3000 F, they were 4 and < 1%, respectively. However, the alloying additions raised the elongation and reduction of area values to 42 to 43 and 52 to 64%, respectively. The differences in fracture appearances of the three alloys are illustrated in Figure 39. The microstructures of undeformed and fractured portions of the tension test specimens are shown in Figure 40.

Summary

The aim of Phase II was to investigate further strengthening of one of the compositions in the W-Ta-Mo-Cb alloy system by carbide dispersions. For this purpose, the effects of small additions of titanium, vanadium, zirconium, and carbon to unalloyed tungsten and the 88W-12Cb alloy were studied.

The primary effort in this phase was expended on the development of suitable melting techniques. As in Phase I, small cylindrical ingots were consumably arc-melted by the multiple electrode technique; the small additions of titanium, vanadium, zirconium, and carbon were made by combining these materials into a composite electrode. Satisfactory ingots of W-12Cb-Zr-C and W-12Cb-V-Zr-C alloys were produced, and vacuum-arc melting of consumable multiple electrodes proved to be an effective method for adding small amounts of reactive metals and carbon to refractory metal alloys. Titanium and vanadium caused difficulties because of their high vapor pressures. Poor recovery and porosity are evidently serious problems with titanium, but the substitution of vanadium resulted in higher recovery and an apparent reduction in porosity.

Billets of the carbon-containing alloys as well as of the 88W-12Cb base composition were impact-extruded to round bars (approximate reduction ratio of 3 to 1).

A comparison of the as-cast, as-extruded, and 3500 F annealed microstructures of the W-12Cb-Zr-C, W-12Cb-V-Zr-C, and 88W-12Cb alloys revealed important recrystallization

and carbide solubility features. The solution-annealed 88W-12Cb alloy was fully recrystallized, whereas the solution-annealed Zr-C-containing and V-Zr-C-containing alloys were less than 50% recrystallized.

The tensile strength of the base composition (88W-12Cb) at 3500 F was 27,000 psi—to our knowledge, higher than any 3500 F tensile strength value reported to date. Moreover, the potent effect of small alloying additions was shown by the 3500 F tensile strengths of 49,000 and 57,000 psi for the W-12Cb-Zr-C and W-12Cb-V-Zr-C alloys, respectively. In addition, the minor alloying additions effected a tremendous improvement in tensile ductility.

Further study of strengthening by dispersion effects should offer a very interesting challenge as well as the most potential for a short-range breakthrough in producing high-strength refractory alloys.

Table I

Tensile Properties at 3000 F of As-Extruded Materials[a]

Alloy	Heat	Nominal Composition (wt. %)				Ultimate Tensile Strength (1000 psi)	Elongation in 0.5 in. (%)	Reduction of Area (%)
		W	Ta	Mo	Cb			
1	56	100.0	-	-	-	14	74	99
2	189T*	75.3	24.7	-	-	64[c]	<1	-
3	192*	50.4	49.6	-	-	64	4	8
4	136F	25.3	74.7	-	-	37	3	1
5	61	-	100.0	-	-	4	118	99
6	141	88.8	-	-	11.2	50	4	<1
7	200F*	68.2	20.6	-	11.2	48[c]	4	-
8	115B	44.4	44.4	-	11.2	54	7	13
9	232B*	20.7	68.1	-	11.2	32	32	60
10	114B	-	88.6	-	11.4	6	102	99
11	82	88.4	-	11.6	-	25	47	93
12	197T*	68.0	20.4	11.6	-	61	~10	13
	197B*					67	16	23
13	103	44.2	44.2	11.6	-	57	5	<1
14	194T*	20.4	67.9	11.7	-	52	30	33
	195B*					55	24	40
15	121	-	88.3	11.7	-	29	29	24
16	143[b]	88.6	-	5.7	5.7	62[c]	~6	-
17	202T*	68.1	20.5	5.7	5.7	54[c]	<1	-
18	101[b]	44.3	44.3	5.7	5.7	50[c]	~2	-
	203					56	<1	<1
19	97[b]	20.6	68.0	5.7	5.7	36	12	15
20	119F	-	88.6	5.7	5.7	18	25	36

[a] An asterisk indicates that data were obtained under the present contract; all other data taken from WADD TR 61-134.

[b] Previously creep-tested for 5 hours at 3000 F under a stress of 7,000 psi.

[c] Fractured at the fillet portion of the specimen.

Table II

Analyses of Electrode Rods

Metal	W	Ta	Mo	Cb
Supplier	GE	Kennametal	GE	Wah Chang
Min. Purity	99.95%	99.9%	99.95%	Avg. As-Cast BHN = 68
Element	w/o[a]	ppm	w/o[b]	ppm
Al	<0.001	---	0.001	<20
B	-	---	-	< 1
C	-	< 10	0.002	30
Ca	<0.001	---	<0.001	---
Cb	-	<300	-	---
Cd	-	---	-	< 5
Co	-	---	-	<20
Cr	<0.001	---	-	<20
Cu	<0.001	---	<0.001	<40
Fe	<0.001	<100	<0.001	<100
H	-	---	-	7
Hf	-	---	-	<80
Mg	0.001	---	<0.001	<10
Mn	<0.001	---	<0.001	<20
Mo	<0.005	---	-	<10
N	-	<10	-	83
Ni	<0.001	---	<0.001	<10
O	-	90	0.050	<50
Pb	-	---	-	<20
Si	<0.001	<100	0.001	<100
Sn	<0.001	---	0.001	<20
Ta	-	---	-	<500
Ti	-	<100	-	<150
V	-	---	-	< 5
W	-	---	-	<200
Zn	-	---	-	<20
Zr	-	---	-	---

[a] Spectrographic analysis of powder lots from which the rods were produced.

[b] Spectrographic analysis of typical powder lots used for producing rods; C and O determined for specific lots used for this study.

Table III

Results of Hardness Tests on Consumable-Electrode Vacuum-Arc-Melted Ingots for Phase I

Alloy	CEVA Heat No.	Average Hardness (DPH)[a]	
		Ingot Top	Ingot Bottom
25W-75Ta	160	336	364
	161	333	369
	162	322	376
	165	367	382
	182	336	361
	183	336	337
88Ta-12Mo	166	337	344
	167	327	350
	168	314	338
	184	341	346
	185	338	325
	186	330	343
44W-44Ta-12Cb	169	419	423
	170	413	409
	171	409	421
	172	399	416
	173	401	419
	174	412	442
	226	430	442
	227	436	460
88W-6Mo-6Cb	178	347	340
	179	341	353
	180	342	-
	181	349	343
	187	350	344
	188	351	348
	216	359	353
	217	350	359
68W-20Ta-12Mo	220	356	378
	221	367	361
	222	352	362
	223	351	377
	224	346	371
	225	-	353

[a] A 10-kg load was used.

Table III (Continued)

Results of Hardness Tests on Consumable-Electrode Vacuum-Arc-Melted Ingots for Phase I

Alloy	CEVA Heat No.	Average Hardness (DPH)[a]	
		Ingot Top	Ingot Bottom
68Ta-20W-12Mo	210	422	417
	211	419	410
	212	407	424
	213	437	435
	214	420	426
	228	426	414
70Ta-19W-11Mo	231	-	417
22W-78Ta	163	323	-
	164	329	367
100W	230	342	353

Table IV

Results of Phase I Extrusion Experiments

Alloy	CEVA Heat No.[a]	Billet			Cropped Extrusion		Approx. Red. of Area[b] (%)	Yield[c] (%)
		Length (in.)	Dia. (in.)	Wt. (g)	Length (in.)	Wt. (g)		
25W-75Ta	160	1.38	1.001	309	3.31	269	64	87
	161	1.54	1.000	344	3.69	305	62	89
	162T	0.93	1.001	207	1.82	163	60	79
	162B	0.96	1.000	216	1.85	167	60	77
	165	1.29	1.002	292	2.80	242	62	83
	182T	1.04	1.000	232	2.09	176	62	76
	182B	1.02	1.000	228	2.29	182	64	80
	183T	0.89	1.000	197	1.99	161	63	82
	183B	0.97	1.000	216	2.12	173	63	80
88Ta-12Mo	166T	0.94	1.000	191	1.99	162	60	85
	166B	1.00	1.000	203	2.06	163	61	80
	167T	0.94	1.001	191	2.26	159	65	83
	167B	0.90	1.000	182	1.75	140	60	77
	168T	0.93	1.000	186	2.19	164	63	88
	168B	1.01	1.000	203	1.99	151	62	74
	184	1.75	1.000	355	2.34	192	59	54
	185	1.25	0.995	350	2.68	325	57	93
	186T	0.93	0.998	186	1.89	151	60	81
	186B	1.00	1.000	201	2.00	162	60	81

[a] "T" denotes top of ingot; "B" denotes bottom of ingot.

[b] Since it was difficult to measure the area of a round-cornered rectangular cross section, the approximate area was calculated from the following formula:

$$A_f = \frac{W_f l_o A_o}{W_o l_f}$$

where A_f = cross-sectional area of the extrusion,
W_f = weight of the extrusion,
l_f = length of the extrusion
l_o = length of the billet,
A_o = cross-sectional area of the billet,
W_o = weight of the billet.

[c] Based on weight.

Table IV (continued)

Results of Phase I Extrusion Experiments

Alloy	CEVA Heat No.[a]	Billet			Cropped Extrusion		Approx. Red. of Area[b] (%)	Yield[c] (%)
		Length (in.)	Dia. (in.)	Wt. (g)	Length (in.)	Wt. (g)		
44W-44Ta-12Cb	169T	0.95	1.000	195	2.02	160	61	82
	169B	0.92	0.999	190	1.94	139	65	73
	170	1.59	1.001	326	3.42	268	62	82
	171	1.55	1.001	320	3.55	283	62	88
	172T	1.00	1.003	207	2.10	162	63	78
	172B	1.00	1.000	207	2.20	172	62	83
	173T	1.04	0.999	215	2.00	160	61	74
	173B	1.06	1.000	217	2.30	178	62	82
	174T	1.06	1.000	219	2.22	175	62	80
	174B	1.07	1.000	222	2.32	166	65	75
	226T	0.89	1.001	179	1.98	141	65	81
	226B	0.96	1.001	195	2.12	148	65	76
	227T	1.01	1.000	215	2.08	148	66	69
	227B	1.07	1.001	227	2.35	165	67	73
88W-6Mo-6Cb	178	1.52	1.001	336	3.40	285	62	85
	179T	0.98	1.000	216	1.94	166	61	77
	179B	0.96	0.999	213	1.63	136	62	64
	180	1.24	0.958	249	2.50	206	59	83
	181T	0.86	1.000	191	1.90	163	61	85
	181B	0.92	1.000	204	1.48	123	63	60
	187	1.11	1.001	245	1.60	139	61	57
	188	1.79	1.000	395	3.83	336	60	85
	216	0.98	0.996	220	2.27	176	66	80
	217	1.22	1.000	263	2.90	224	64	85
68W-20Ta-12Mo	220T	0.84	1.000	180	1.44	111	63	62
	220B	0.75	1.000	158	1.58	118	64	75
	221T	0.98	1.000	205	2.14	160	64	78
	221B	0.98	1.000	210	1.98	147	65	70
	222T	0.86	0.997	184	1.88	140	65	76
	222B	0.87	1.000	188	2.04	151	66	80
	223T	1.04	1.000	225	2.23	159	67	71
	223B	1.19	1.000	257	2.22	170	66	65
	224T	0.96	1.000	205	2.59	186	66	91
	224B	1.00	1.000	212	2.28	172	64	81
	225T	0.91	1.001	199	2.00	174	60	87
	225B	0.92	1.001	200	2.30	173	65	87

Table IV (continued)

Results of Phase I Extrusion Experiments

Alloy	CEVA Heat No.[a]	Billet			Cropped Extrusion		Approx. Red. of Area[b] (%)	Yield[c] (%)
		Length (in.)	Dia. (in.)	Wt. (g)	Length (in.)	Wt. (g)		
68Ta-20W-12Mo	210T	1.05	1.000	215	2.50	171	67	80
	210B	1.12	1.000	226	2.92	198	66	88
	211T	1.23	1.000	251	3.28	224	67	89
	211B	1.21	1.000	245	2.70	192	65	78
	212T	1.14	1.000	233	2.82	194	66	83
	212B	1.10	1.000	224	2.78	187	67	83
	213T	1.16	0.999	236	3.00	206	66	87
	213B	1.26	1.000	257	3.30	224	67	87
	214T	1.16	0.997	234	2.98	208	66	88
	214B	1.20	0.999	243	3.07	209	66	86
	228T	1.00	1.000	203	2.41	168	65	83
	228B	1.02	1.000	201	2.37	162	65	81
70Ta-19W-11Mo	231T	1.08	1.000	219	3.08	188	70	86
	231B	0.96	1.002	198	3.45	170	76	86
22W-78Ta	163T	0.91	1.001	202	1.98	167	62	83
	163B	0.98	1.001	218	1.60	146	59	67
	164T	1.02	1.002	227	2.04	184	59	81
	164B	1.04	1.001	232	2.43	202	63	87
100W	230T	1.10	1.000	244	2.70	225	62	92
	230B	1.10	1.000	267	3.04	241	67	90

Table V

Rolling Schedules for Reducing Sheet Bars to Intermediate-Gauge Sheets

Alloy	Rolling Pass	Approximate Temperature[a] of Sample Before Rolling (°F)	Roll Set-Down (0.001 inch)
75Ta-25W	1	3500	25
	2	3400-3500	25
	3	3300-3400	50
	4	3300-3400	50
	5	3100-3200	50
	6	3100-3200	50
	7	3100-3200	50
88Ta-12Mo	1	3400	25
	2	3300-3400	25
	3	3300-3400	50
	4	3100-3200	50
	5	3100-3200	50
	6	3000-3100	50
	7	3000-3100	50
100W	1	3200	25
	2	3100-3200	50
	3	3000-3100	50
	4	2900-3000	50
	5	2800-2900	50
	6	2800-2900	50
	7	2700-2800	50

[a]Uncorrected optical pyrometer reading.

Table VI

Rolling Schedules for Reducing Intermediate-Gauge Sheets to Final-Gauge Sheets

Alloy	Rolling Pass	Roll Set-Down (0.001 inch)	Approximate Temperature[a] (°F) before Rolling a Sample That Requires			
			5 Passes	4 Passes	3 Passes	2 Passes
75Ta-25W	1	25	3200	3200	3200	3000
	2	25	3000	3000	2800	2600
	3	25	3000	2800	2600	--
	4	25	2800	2600	--	--
	5	25	2600	--	--	--
88Ta-12Mo	1	25	3000	3000	3000	2800
	2	25	3000	2800	2800	2600
	3	25	2800	2800	2600	--
	4	25	2800	2600	--	--
	5	25	2600	--	--	--
100W	1	25	3000	2800	2800	2600
	2	25	2800	2800	2600	2400
	3	25	2800	2600	2400	--
	4	25	2600	2400	--	--
	5	25	2400	--	--	--

[a] Uncorrected optical pyrometer reading.

Table VII

Results of Rolling Experiments on the 75Ta-25W Alloy

CEVA Heat No.	Code	Sheet Bar Thickness (in.)	Int.-Gauge Sheet Thickness (in.)	Final Gauge Sheet Thickness (in.)	Reduction in Thickness (%) Final-Gauge Sheet[c]				
					Sheet Bar to Int. Gauge	Int. to Final Gauge	Total	Length (in.)	Condition
160	160-1	0.480	0.225	0.040	32	82	92	6.3	Poor
	160-2	0.480	0.225	0.065	32	71	86	4.8	Good
162	162T	0.470	0.175	0.076	63	57	84	5.1	Very Good
	162B	0.417	0.306	0.070	27	77	83	3.7	Fair
163[a]	163B	0.440	0.185	0.051	58	72	88	5.0	Poor
164[b]	164T	0.460	0.121	0.072	74	41	84	4.3	Good
	164B	0.490	0.137	0.071	72	48	86	6.8	Very Good
165	165-1	0.450	0.140	0.069	69	51	85	4.3	Good
	165-2	0.450	0.140	0.063	69	55	86	3.6	Good
182	182T	0.470	0.124	0.074	73	40	84	4.5	Excellent
	182B-1	0.450	0.125	0.069	72	45	85	5.7	Good
	182B-2	0.450	0.125	0.069	72	45	85	2.3	Very Good
183	183T	0.400	--[b]	0.060	--[b]	--[b]	85	7.7	Excellent
	183B	0.375	0.135	0.065	64	48	81	5.5	Excellent

[a] Off-composition heat (78Ta-22W) melted and extruded for preliminary rolling trials.

[b] Rolled directly from sheet bar to final-gauge sheet.

[c] Average width = 0.7 inch.

Table VIII

Results of Rolling Experiments on the 88Ta-12Mo Alloy and Unalloyed Tungsten

CEVA Heat No.	Code	Sheet Bar Thickness (in.)	Int.-Gauge Sheet Thickness (in.)	Final Gauge Sheet Thickness (in.)	Reduction in Thickness (%) Final-Gauge Sheet[c]				
					Sheet Bar to Int. Gauge	Int. to Final Gauge	Total	Length (in.)	Condition
166	166T	0.480	0.161	0.085	67	47	82	4.0	Fair
	166B	0.480	0.137	0.074	72	46	85	6.2	Good
167	167T-1	0.470	0.220	0.078	53	65	83	3.5	Fair
	167T-2	0.470	0.220	0.080	53	64	83	3.2	Good
	167B	0.480	0.110	0.048	77	56	90	5.9	Fair
168	168T	0.470	0.160	0.068	66	43	80	4.2	Excellent
	168B	0.450	--[a]	0.086	--[a]	--[a]	81	9.6	Excellent
184	184-1	0.480	0.135	0.070	72	48	85	3.7	Good
	184-2	0.480	0.135	0.074	72	55	90	4.6	Fair
185	185-1	0.420	0.170	0.075	60	56	82	4.7	Excellent
	185-2	0.420	0.170	0.077	60	55	82	5.8	Good
186	186T	0.480	0.130	0.080	73	39	83	4.8	Very Good
	186B	0.490	0.135	0.092	73	32	81	4.4	Very Good
230[b]	230T	0.390	0.104	0.069	73	34	83	5.6	Good
	230B	0.385	0.130	0.080	66	39	79	7.3	Good

[a] Rolled directly from sheet bar to final gauge sheets in previous rolling experiments.

[b] Unalloyed tungsten.

[c] Average width = 0.7 inch.

Table IX

Summary of Melting, Extrusion, and Rolling for Phase I

Alloy	Ingot No.	Billets Extruded to Sheet Bars		Sheet Bars Rolled to Intermediate Gauge			Intermediate-Gauge Sheets Rolled to Final Gauge		
		Available	Successful	Available	Successful	Unsuccessful	Available	Successful	Unsuccessful
25W-75Ta	160	1	1	1	1	-	2	2	-
	161	1	1	1	-	1	-	-	-
	162	2	2	2	2	-	2	2	-
	163[a]	2	2	2	2	-	2	1	-
	164[a]	2	2	2	2	-	2	2	-
	165	1	1	1	1	-	2	2	-
	182	2	2	2	2	-	3	3	-
	183	2	2	2	2	-	2	2	-
88Ta-12Mo	166	2	2	2	2	-	2	2	-
	167	2	2	2	2	-	3	3	-
	168	2	2	2	2	-	2	2	-
	184	1	1	1	1	-	2	2	-
	185	1	1	1	1	-	2	2	-
	186	2	2	2	2	-	2	2	-
44W-44Ta-12Cb	170	1	1	2	-	2	-	-	-
	171	1	1	2	-	2	-	-	-
	172	2	2	2	-	2	-	-	-
	173	2	2	2	-	2	-	-	-
	174	2	2	2	-	2	-	-	-
	226	2	2	2	-	1	-	-	-
	227	2	2	2	-	-	-	-	-
88W-6Mo-6Cb	178	1	1	1	-	1	-	-	-
	179	2	2	2	-	2	-	-	-
	180	1	1	2	-	2	-	-	-
	181	2	2	2	-	2	-	-	-
	187	1	1	1	-	1	-	-	-
	188	1	1	2	-	2	-	-	-
	216	1	1	1	-	1	-	-	-
	217	1	1	2	-	2	-	-	-
68W-20Ta-12Mo	220	2	2	2	-	2	-	-	-
	221	2	2	2	-	1	-	-	-
	222	2	2	2	-	1	-	-	-
	223	2	2	2	-	2	-	-	-
	224	2	2	2	-	1	-	-	-
	225	2	2	2	-	-	-	-	-
68Ta-20W-12Mo	210	2	2	2	-	2	-	-	-
	211	2	2	2	-	1	-	-	-
	212	2	2	2	-	-	-	-	-
	213	2	2	2	-	1	-	-	-
	214	2	2	2	-	2	-	-	-
	228	2	2	2	-	1	-	-	-
	231[b]	2	2	2	-	-	-	-	-
100W	230	2	2	2	2	-	2	2	-

[a]Off-composition heat (78Ta-22W) melted and extruded for preliminary rolling trials.

[b]Off-composition heat (70Ta-19W-11Mo) melted and extruded for making tensile grip pins.

Table X

Results of Recrystallization Studies on the 75Ta-25W Alloy

Annealing Temperature[a] (°F)	Heat 160-1		Heat 162B		Heat 183T	
	Recrystallization[b] (%)	Hardness[c] (DPH)	Recrystallization[b] (%)	Hardness[c] (DPH)	Recrystallization[b] (%)	Hardness[c] (DPH)
As-rolled	0	461	0	461	0	440
1900	0	433	---	---	0	433
2100	0	429	---	---	0	416
2300	0	433	---	---	0	413
2500	0	413	---	---	0	413
2700	0	433	0	417	0	409
2750	30	394	---	---	0	387
2800	40	425	15	405	35	383
2850	60	387	50	401	50	376
2900	70	409	80	383	80	366
2950	80	394	---	---	90	387
3000	100	---	100	383	100	360
3100	100	---	100	383	100	370

[a] Annealing time was 1 hour.

[b] Metallographically determined by visual examination at 100X.

[c] A 10-kg load was used.

Table XI

Results of Recrystallization Studies
on the 88Ta-12Mo Alloy

Annealing Temperature[a] (°F)	Heat 184-2		Heat 186T	
	Recrystallization[b] (%)	Hardness[c] (DPH)	Recrystallization[b] (%)	Hardness[c] (DPH)
As-rolled	0	450	0	426
1900	0	420	0	401
2100	0	413	0	409
2300	0	409	0	394
2500	0	390	0	390
2550	20	380	35	370
2600	40	387	40	376
2650	70	360	70	370
2700	100	351	100	350
2800	100	360	100	344
2900	100	344	100	344
3000	100	342	100	344

a Annealing time was 1 hour.

b Metallographically determined by visual examination at 100X.

c A 10-kg load was used.

Table XII

Results of Recrystallization Studies on Unalloyed Tungsten

Annealing Temperature[a] (°F)	Recrystallization[b] (%)	Hardness[c] (DPH)
As-rolled	0	464
2100	0	452
2250	0	434
2300	10	438
2350	25	421
2400	40	390
2450	60	382
2500	70	377
2550	80	374
2600	90	356
2650	100	342
2700	100	345
2900	100	342
3100	100	339

a Annealing time was 1 hour.

b Metallographically determined by visual examination at 100X.

c A 10-kg load was used.

Table XIII

Summary of Recrystallization Temperatures of Selected Warm-Worked Refractory Metals and Alloys

Composition	Recrystallization Temperature[a] (°F)
Mo[b]	2000
Ta[c]	2000
W	2425
Ta-10W[d]	2450
Ta-20W[d]	2800
Ta-25W	2850
Ta-5Mo[c]	2450
Ta-12Mo	2600

[a]Based on 50% recrystallization.

[b]R. I. Jaffee and D. J. Maykuth, "Refractory Materials," DMIC Memorandum 44, February 26, 1960, p. 17.

[c]F. F. Schmidt, "Tantalum and Tantalum Alloys," DMIC Report 133, July 25, 1960, p. 304.

[d]P. P. Crimmins and C. W. Heimlich, "Spinning of Refractory Alloys," Metal Progr. pp 67-72 (December 1962).

Table XIV

4T Bend Transition Data on Sheet Material

Alloy	Test[a] Temp. (°F)	Max. Dial[b] Deflection with Load (in.)	Final Bend Angle Unloaded (deg)	Sample Condition after Test
100W	250	0.008	4	Cracked
	350	0.047	11	Cracked
	400	0.009	5	Cracked
	450	0.012	7	Cracked
	450	0.270	98	Cracked
	500	0.280	99	Uncracked
	550	0.270	90	Uncracked
75Ta-25W	250	0.030	9	Cracked
	350	0.060	22	Cracked
	400	0.197	27	Cracked
	450	0.160	55	Cracked
	450	0.280	96	Uncracked
	500	0.150	58	Cracked
	500	0.280	97	Uncracked
	600	0.280	95	Uncracked
88Ta-12Mo	200	0.010	1	Cracked
	300	0.050	27	Cracked
	300	0.280	98	Uncracked
	350	0.150	60	Cracked
	400	0.280	100	Uncracked
	450	0.210	81	Cracked
	500	0.280	103	Uncracked

[a]Specimens held at temperature for 15 min prior to testing.

[b]Bend angle under load could not be calculated from dial deflection since sample bend apex did not remain in contact with punch throughout bend test.

Table XV

Results of Vacuum Tension Tests on Sheet Specimens of 75Ta-25W, 88Ta-12Mo, and Unalloyed Tungsten

Material	Condition	Test Temperature (°F)	Tensile Strength (1000 psi)	Elongation in 0.6 in. (%)
75Ta-25W	Stress-relieved at 2300 F for 1 hour	2500	83	8
		3000	38	10
		3500	21	12
88Ta-12Mo	Stress-relieved at 2300 F for 1 hour	3000	23	25
		3500	12	35
100W	Stress-relieved at 1900 F for 1 hour	3000	13	93
		3000	18	82
		3500	10	97

Table XVI

Results of Phase II Extrusion Experiments

Alloy	CEVA Heat No.[a]	Billet			Cropped Extrusion			Red. of Area (%)	Red. Ratio	Yield[b] (%)
		Length (in.)	Diameter (in.)	Weight (g)	Length (in.)	Diameter (in.)	Weight (g)			
88W-12Cb	215T	0.74	0.994	157	2.04	0.55	140	69	3.3	89
	215B	0.64	0.995	142	1.69	0.55	114	69	3.3	80
	218T	1.09	0.999	240	2.80	0.56	188	69	3.2	78
	218B	1.01	0.999	224	2.81	0.58	188	66	3.0	84
	219T	0.63	1.000	137	1.23	0.60	91	64	2.8	66
	219B	0.71	1.000	152	1.80	0.58	121	66	3.0	80
W-12Cb-Zr-C	207T	0.77	1.000	171	1.92	0.58	131	66	3.0	77
	207B	0.71	1.000	157	1.82	0.55	120	70	3.3	79
W-12Cb-V-Zr-C	229	1.32	1.001	284	3.81	0.58	249	66	3.0	88
	233	1.03	0.960	187	-	-	-	-	-	0

[a] "T" denotes top of ingot; "B" denotes bottom of ingot.

[b] Based on weight.

Table XVII

As-Extruded Tensile Properties of Phase II Alloys in Vacuum

Alloy	CEVA Heat No.	Test Temp. (°F)	Tensile Strength (1000 psi)	Elong. in 0.5 Inch (%)	Red. of Area (%)
88W-12Cb[a]	141	3000	50	4	<1
88W-12Cb	218B	3500	27	<1	<1
W-12Cb-Zr-C	207T	3500	49	42	64
W-12Cb-V-Zr-C[b]	229	3500	57	43	52

[a] Data taken from WADD TR61-134.

[b] Retested; in the initial test, the specimen grips failed when the stress on the specimen was 55,000 psi.

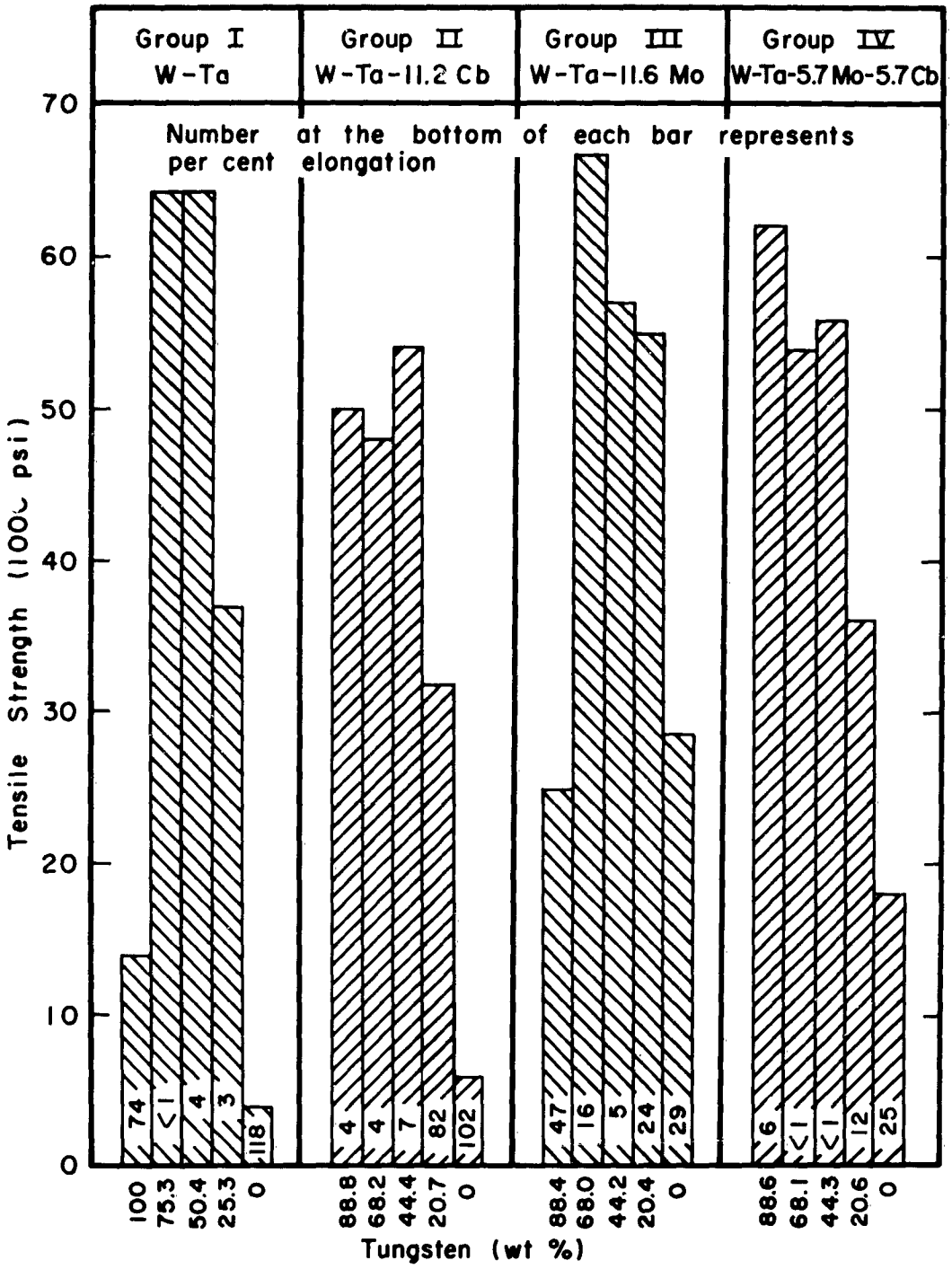
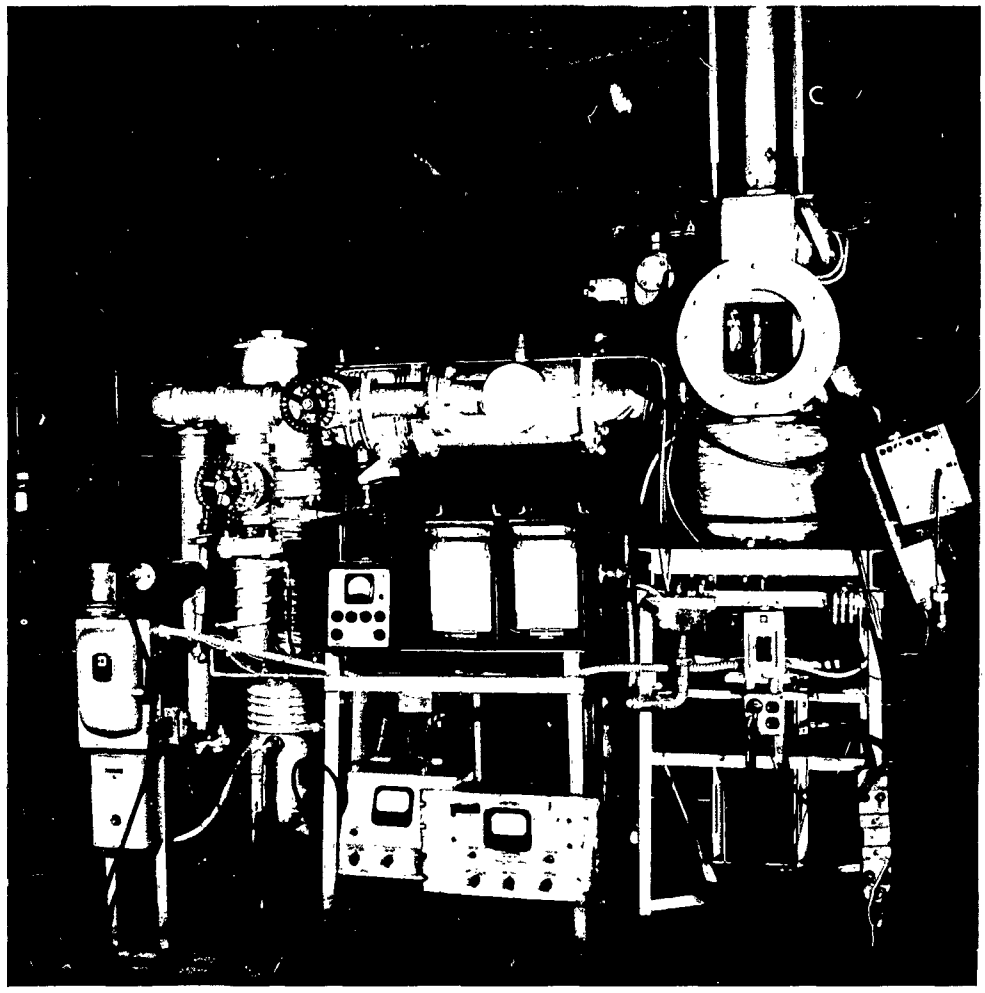


Figure 1

As-Extruded Tensile Strength at 3000 F as a Function of Composition



1099-60

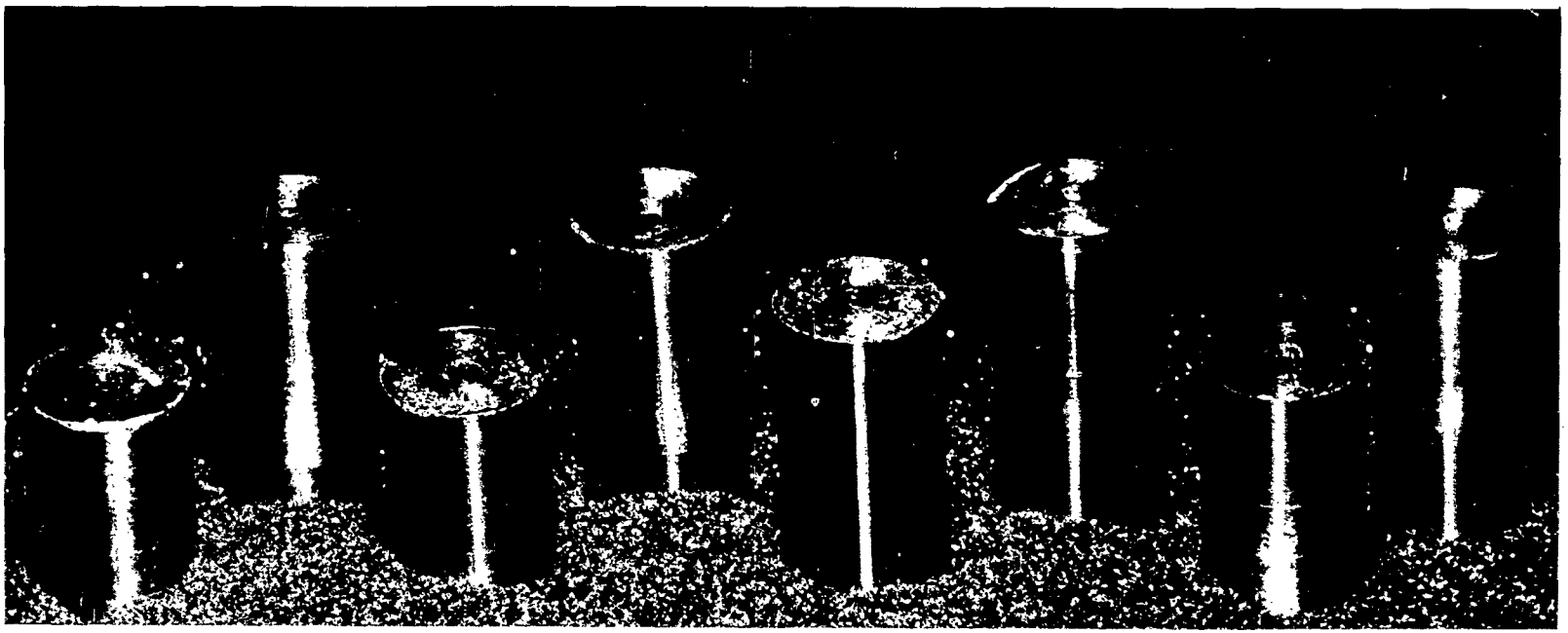
Figure 2

Over-All View of the Melting Equipment

412-62

Figure 3

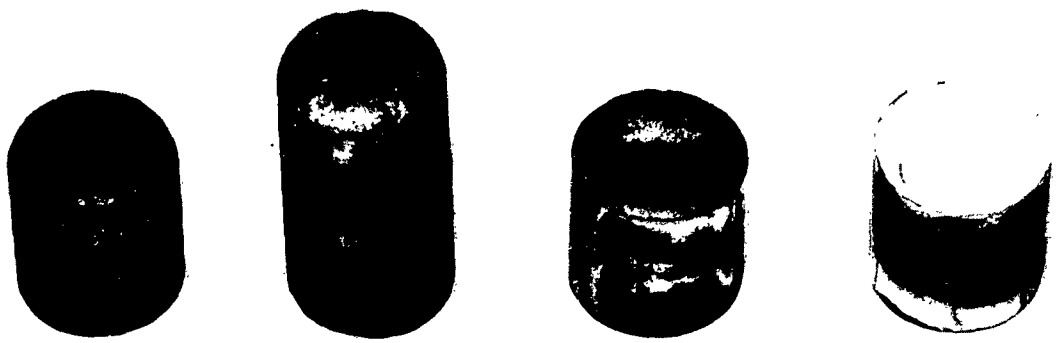
Ingots of the 68Ta-20W-12Mo Alloy
(3/4 Actual Size)



626-62

Figure 4

Ingots of the 68W-20Ta-12Mo and the 68Ta-20W-12Mo Alloys after Lathe Turning and Cylindrically Grinding (Actual Size)



1 2 3 4

Figure 5

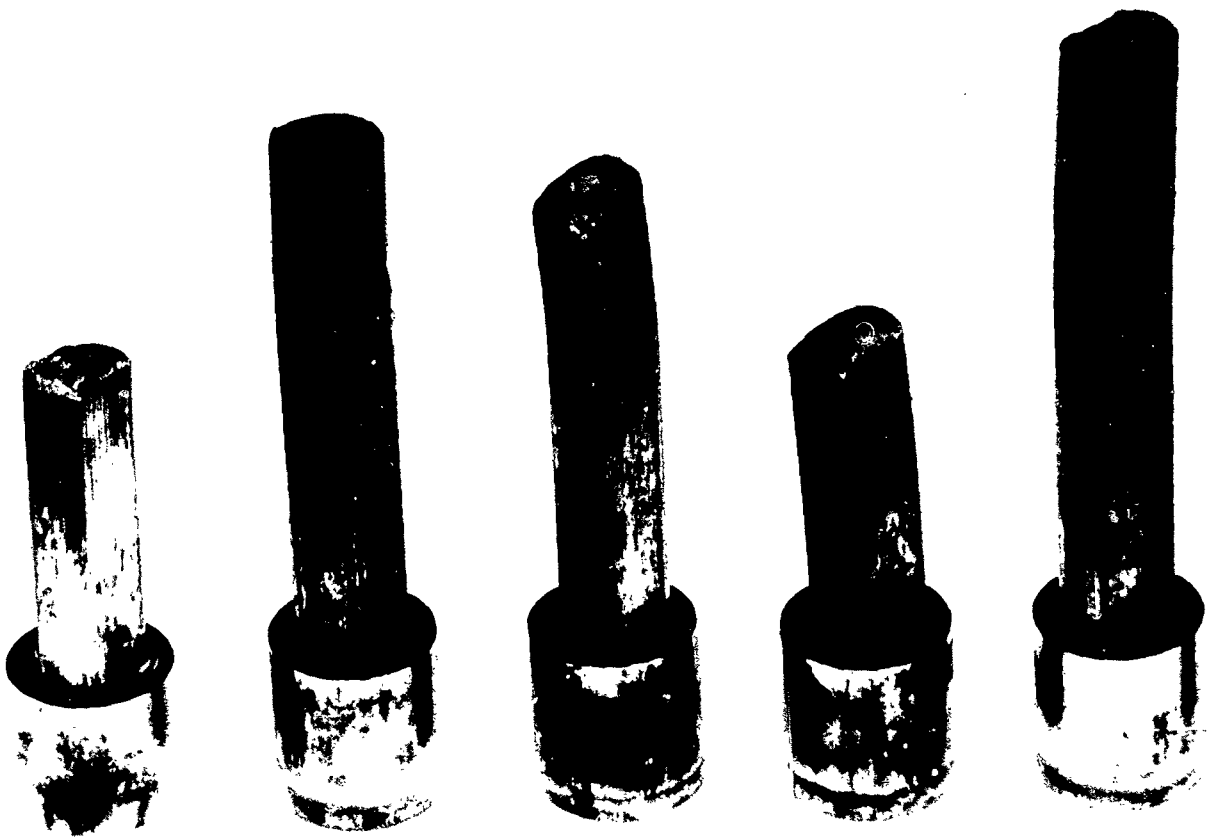
Typical Extrusion Billets and Die Inserts (Actual Size)

(1) 44W-44Ta-12Cb (molybdenum-coated)

(2) 88W-6Mo-6Cb (molybdenum-coated)

(3) Die insert sprayed with molybdenum and tungsten

(4) Die insert sprayed with molybdenum and zirconia



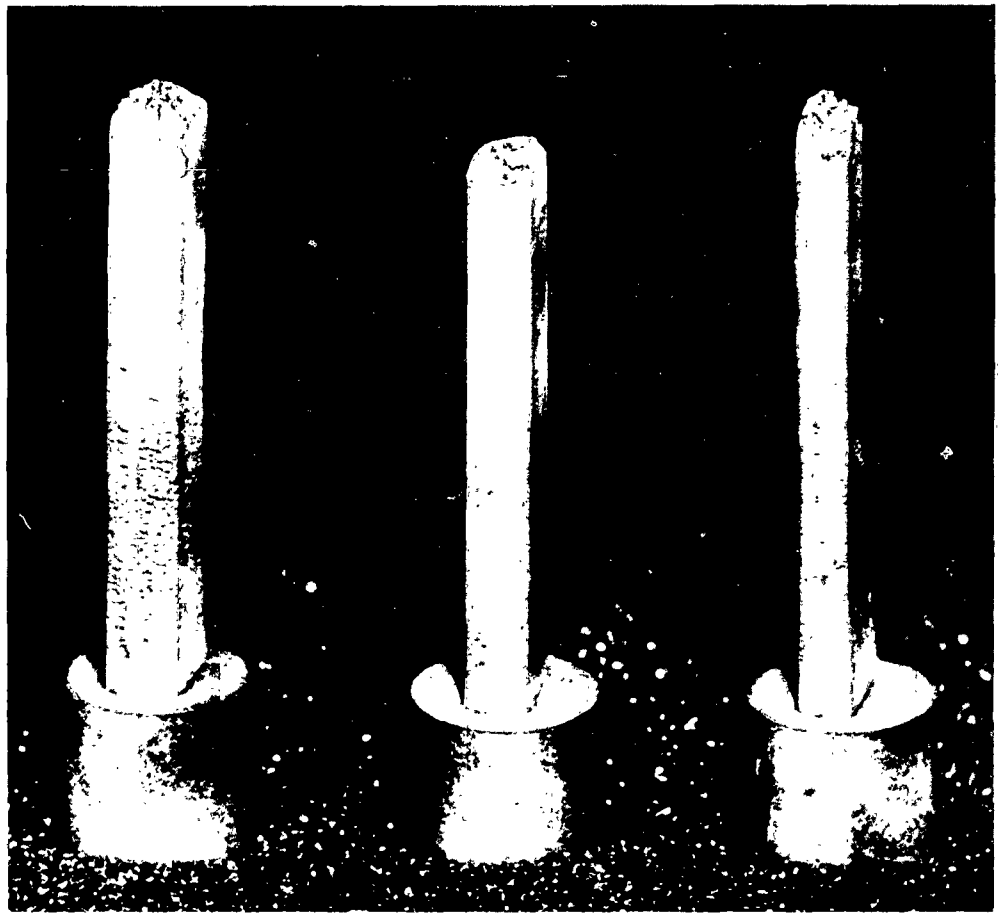
1000-61A

1 2 3 4 5

Figure 6

Typical Sheet-Bar Extrusions (Actual Size)

(1) 100W
(2) 75Ta-25W
(3) 75Ta-25W
(4) 44W-44Ta-12Cb
(5) 88Ta-12Mo



68Ta-20W-12Mo
Heat 211T

70Ta-19W-11Mo
Heat 231T

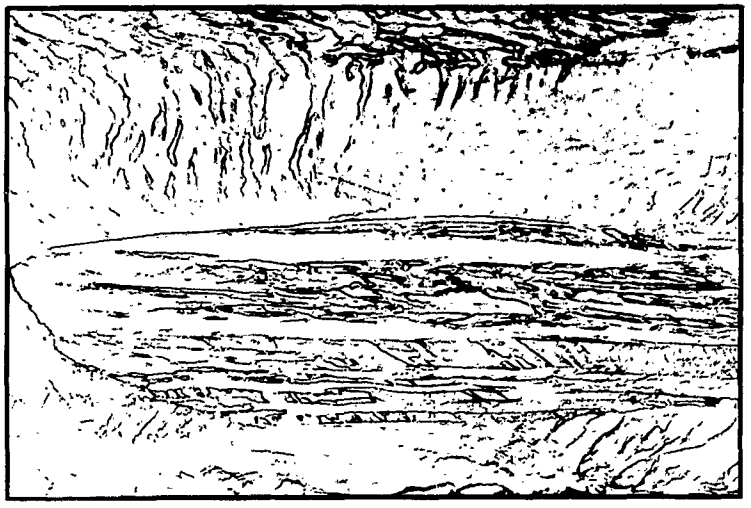
70Ta-19W-11Mo
Heat 231B

Figure 7

Sheet-Bar Extrusions with Reductions of Area of 67, 70, and 76% (Actual Size)

1067-62

75Ta-25W Heat 183T



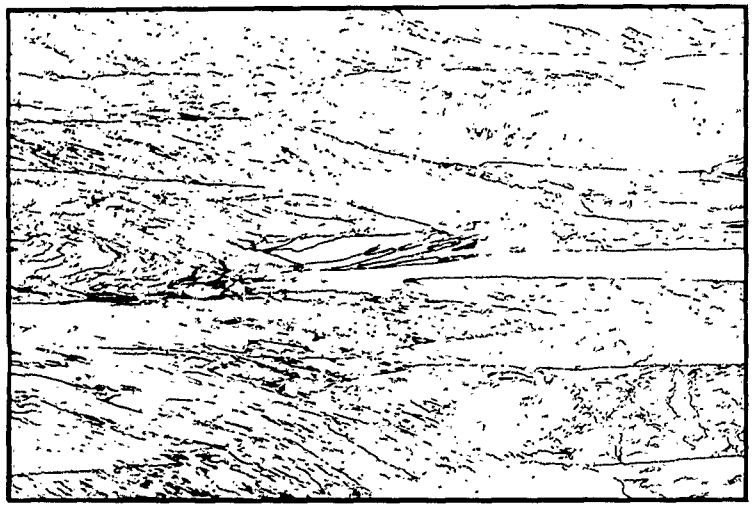
1143-62

88Ta-12Mo Heat 186T

Figure 8

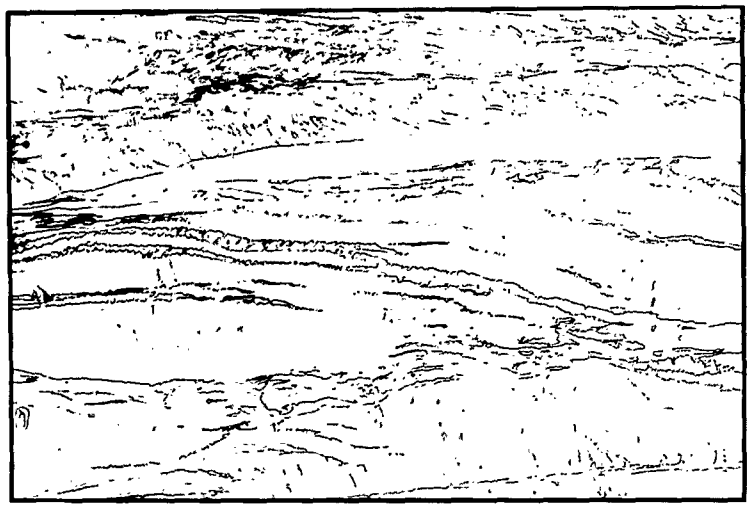
Representative As-Extruded Microstructures of the 75Ta-25W and 88Ta-12Mo Alloys (100X)

Etchant: Mixed Acids



1081-62

68W-20Ta-12Mo Heat 221B



1131-62

68Ta-20W-12Mo Heat 210T

Figure 9

Representative As-Extruded Microstructures of the 68W-20Ta-12Mo and 68Ta-20W-12Mo Alloys (100X)

Etchant: Mixed Acids

1056-62

44W-44Ta-12Cb Heat 170

1068-62

88W-6Mo-6Cb Heat 187

Figure 10

Representative As-Extruded Microstructures of the 44W-44Ta-12Cb and 88W-6Mo-6Cb Alloys (100X)

Etchant: Mixed Acids

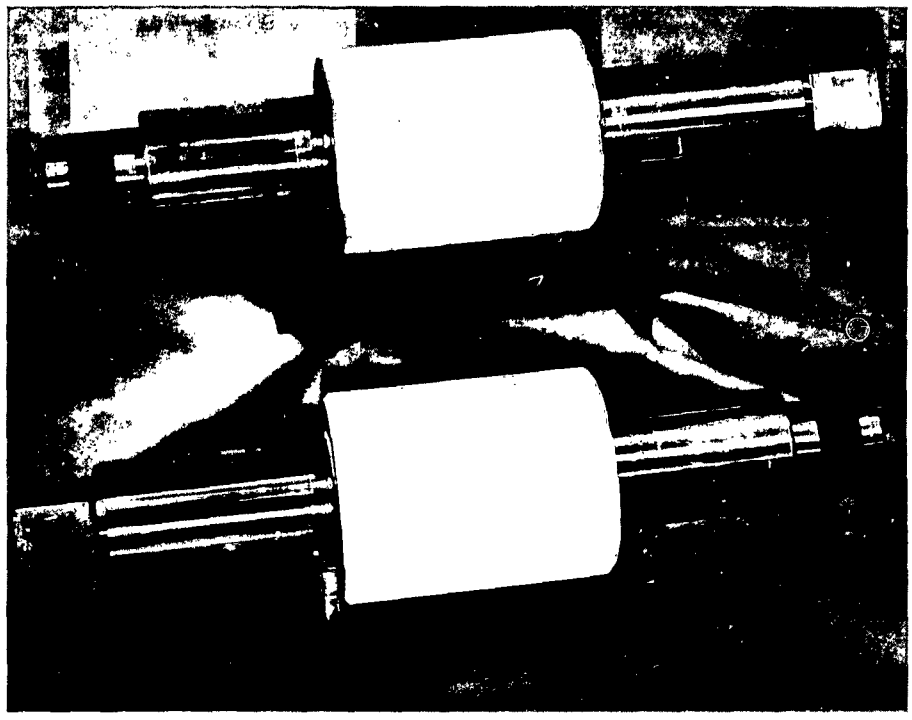
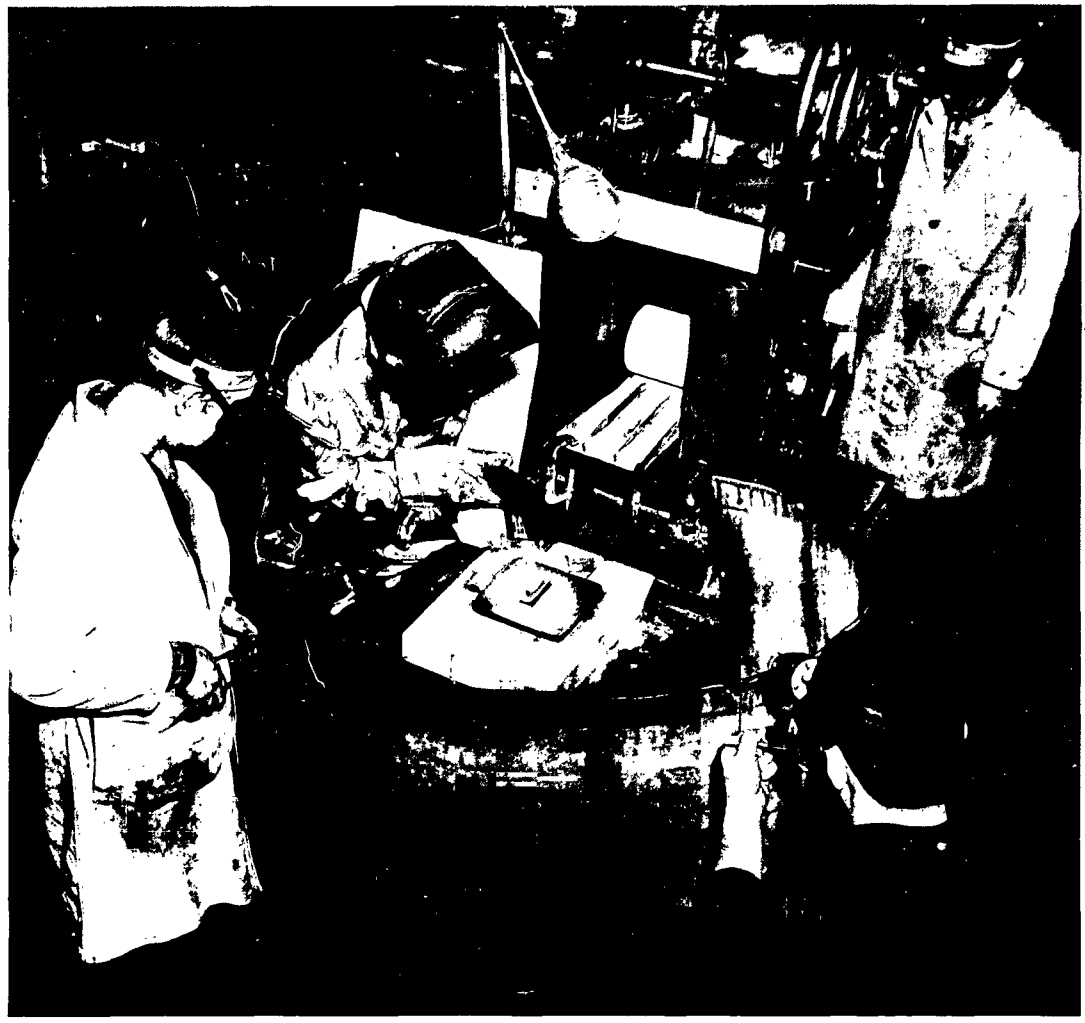


Figure 11

Zirconia-Coated Rolls used in the Stanat Mill (1/5 Actual Size)



177-62

Figure 12

Setup for Plasma Heating and Rolling

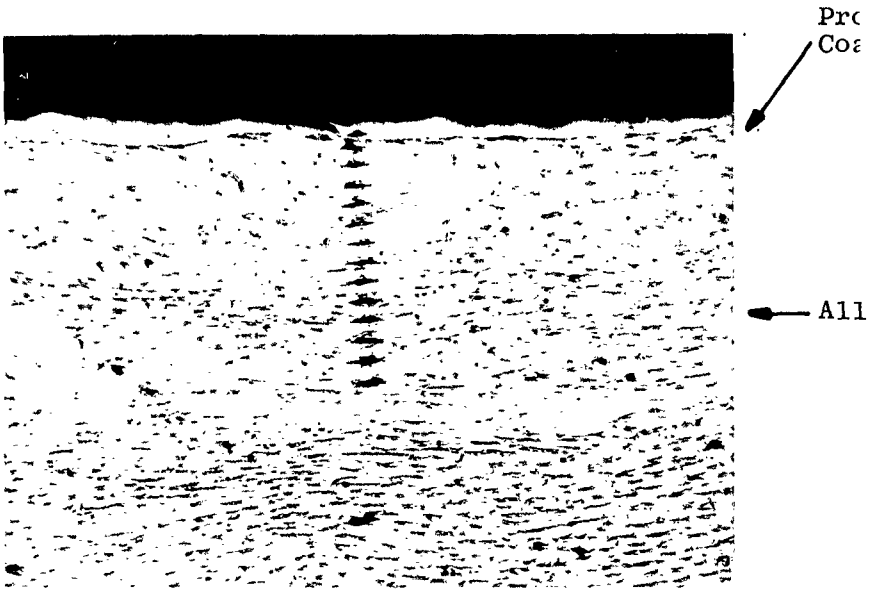
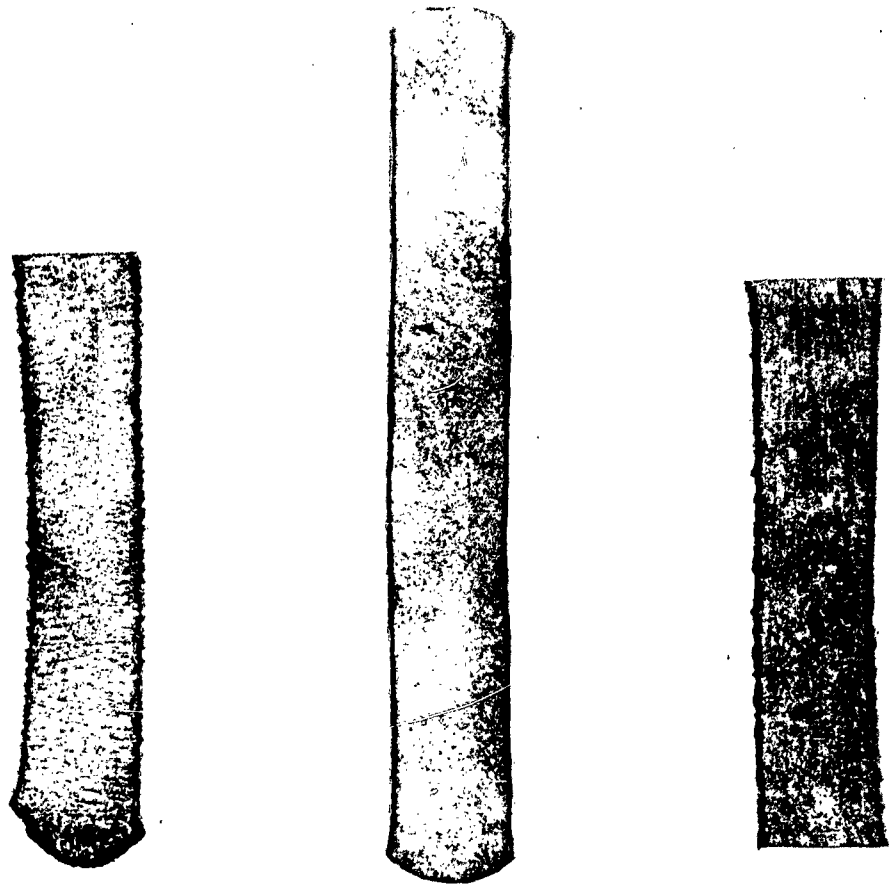


Figure 13

75Ta-25W Alloy (CEVA Heat 165) after 69% Reduction in Thickness by Hot Rolling (100X)

Etchant: 50 cc Lactic Acid; 10 cc HNO_3; 1 cc HF

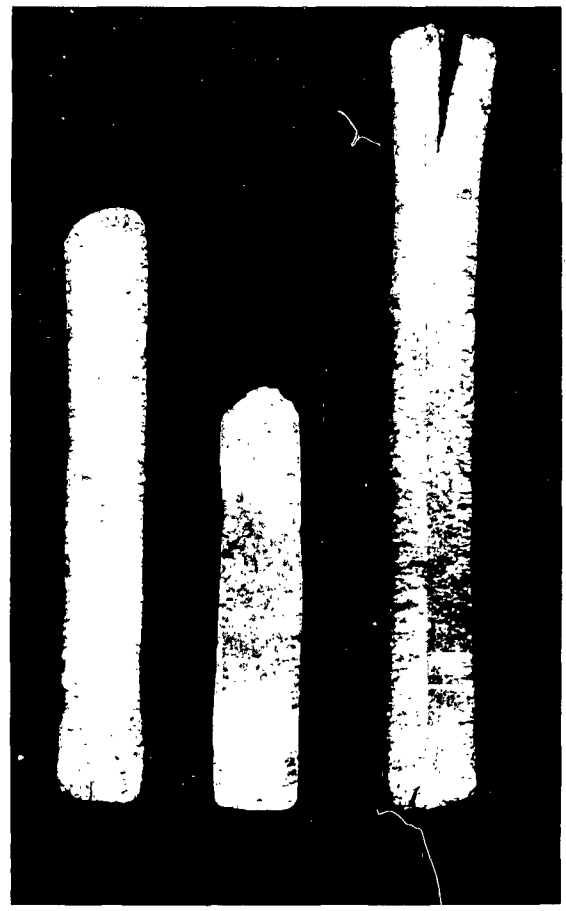


206-62

75Ta-25W	88Ta-12Mo	88Ta-12Mo
Heat 182T	Heat 185	Heat 186B

Figure 14

Sheet Bars Hot-Rolled to Intermediate-Gauge Sheet (Actual Size)

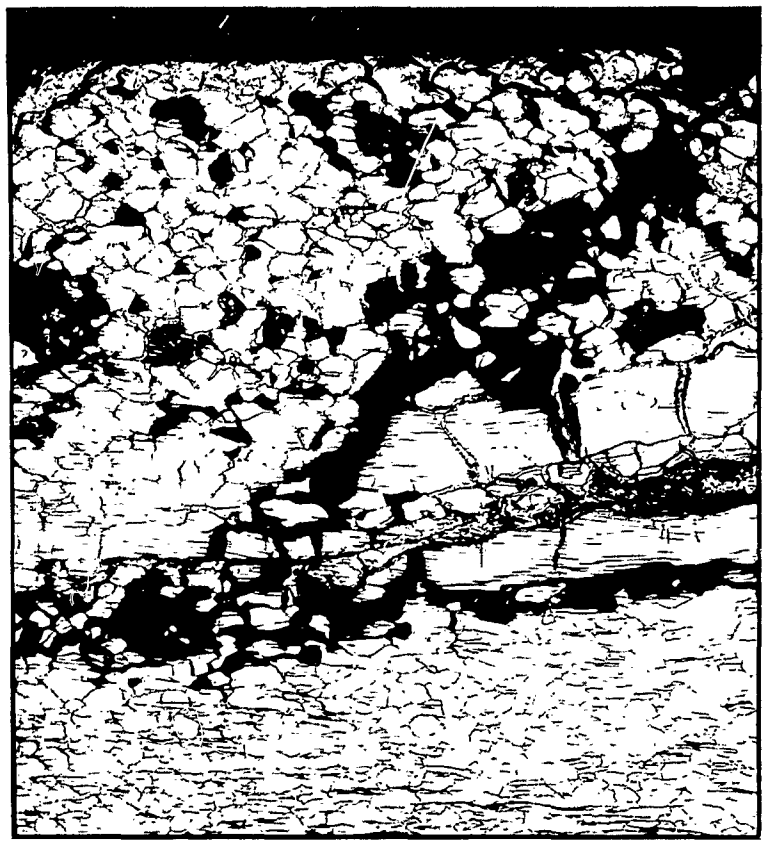


489-62

Figure 15

Sheet Samples of the 75Ta-25W Alloy
(3/4 Actual Size)

Left to Right: Heats 162T, 183T-2, and 164B

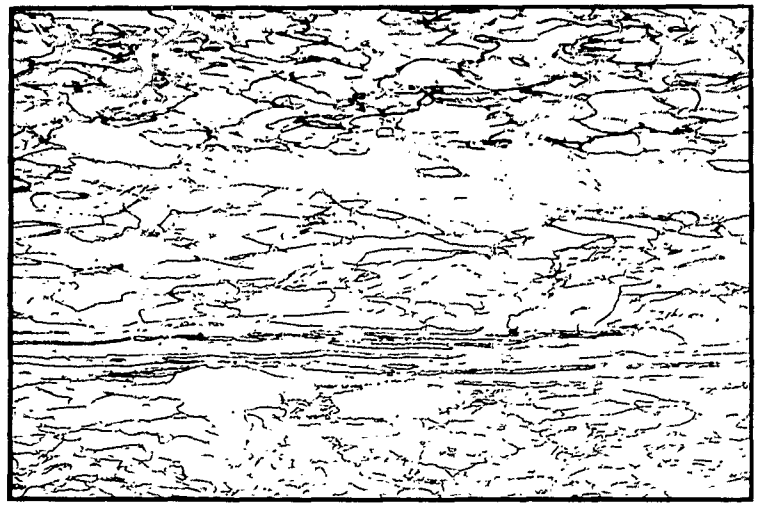


1074-62

Figure 16

Microstructure of a Sheet-Bar Sample of the 44W-44Ta-12Cb Alloy (Heat 173T) that Failed during Hot Rolling (100X)

Etchant: Mixed Acids



1049-62

Intermediate Gauge Heat 162T

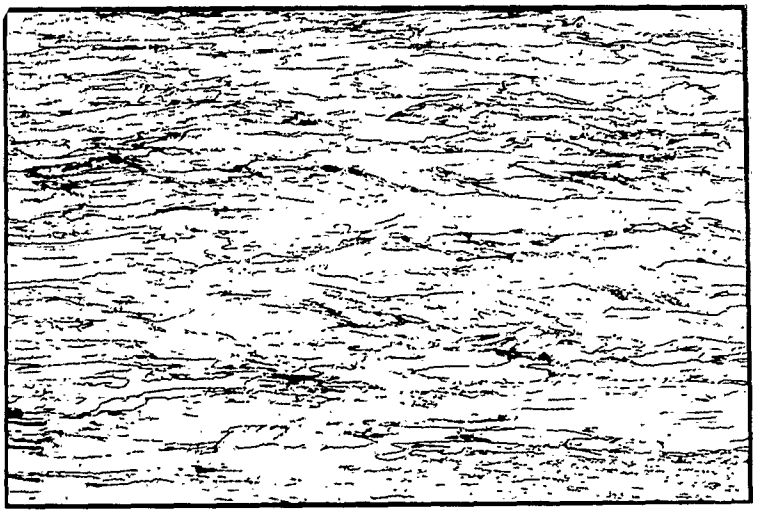
1048-62

Final Gauge Heat 182T

Figure 17

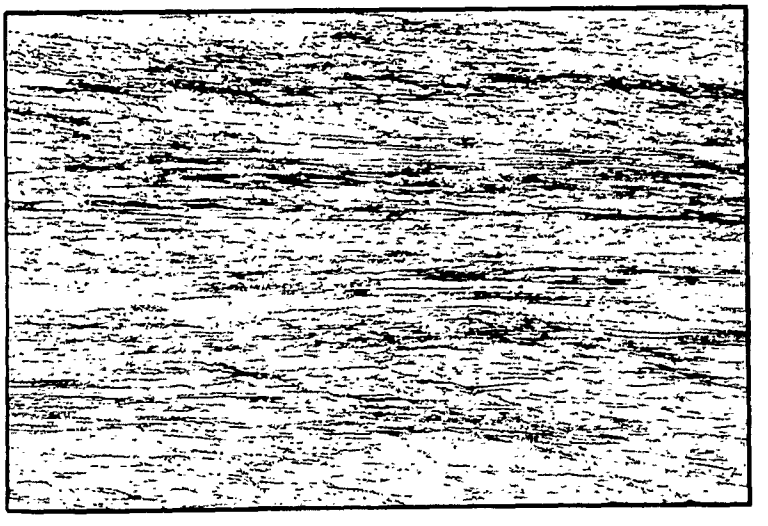
Microstructures of Intermediate- and Final-Gauge Sheets of the 75Ta-25W Alloy (100X)

Etchant: Mixed Acids



1044-62

Intermediate Gauge Heat 166B



1145-62

Final Gauge Heat 186T

Figure 18

Microstructures of Intermediate- and Final-Gauge Sheets of the 88Ta-12Mo Alloy (100X)

Etchant: Mixed Acids

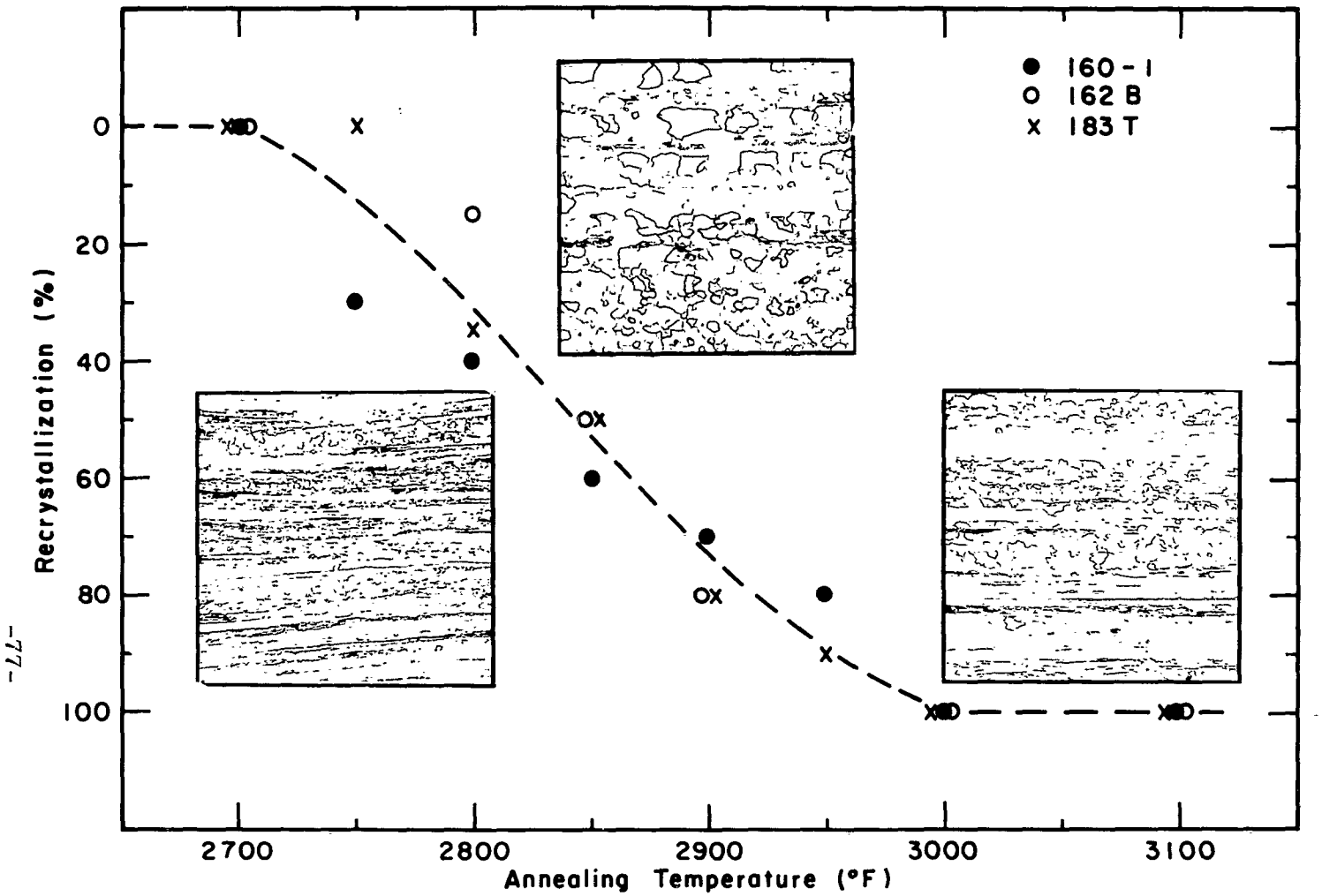


Figure 19

Recrystallization Behavior of the 75Ta-25W Alloy (1-Hour Annealing Time)

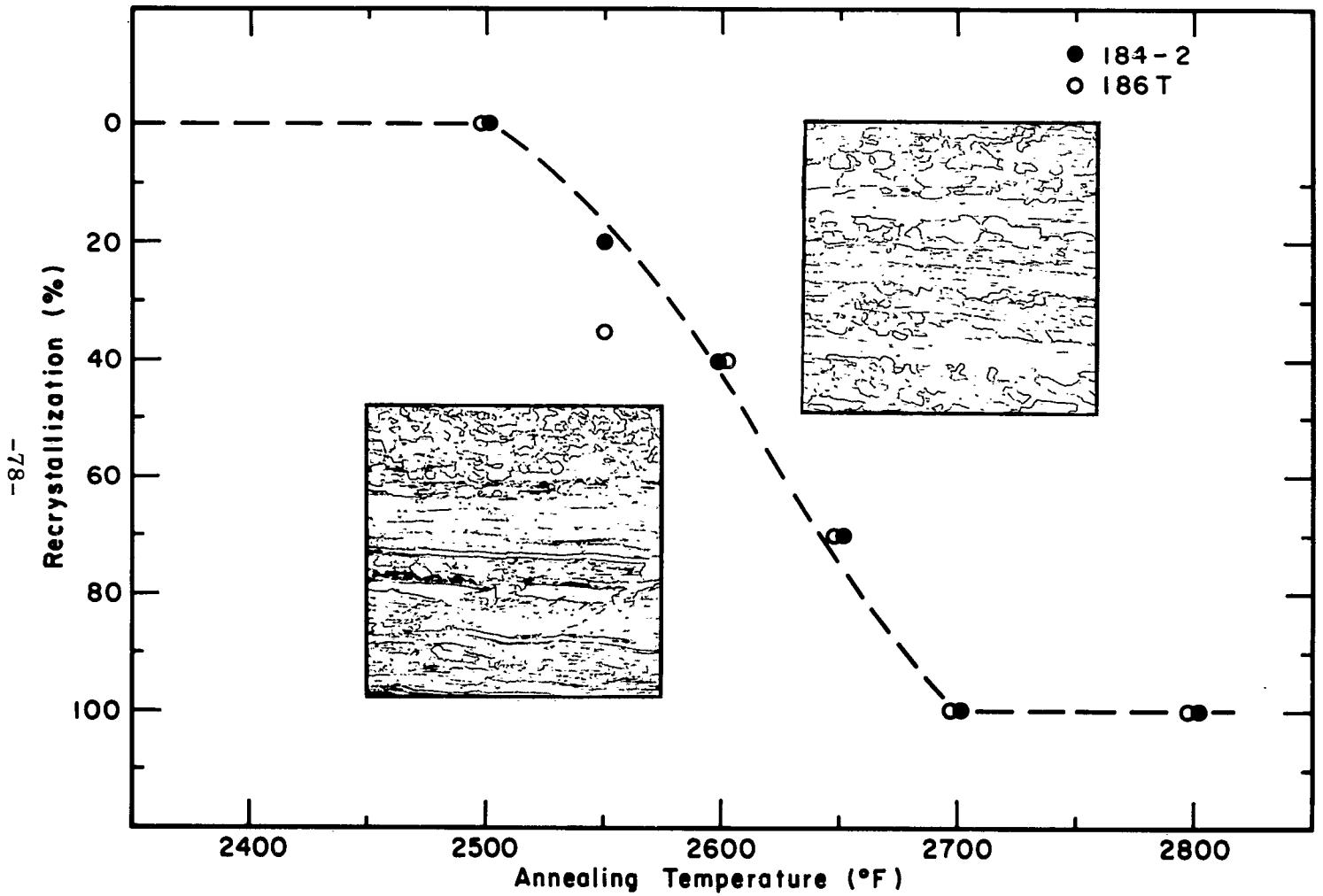


Figure 20

Recrystallization Behavior of the 88Ta-12Mo Alloy (1-Hour Annealing Time)

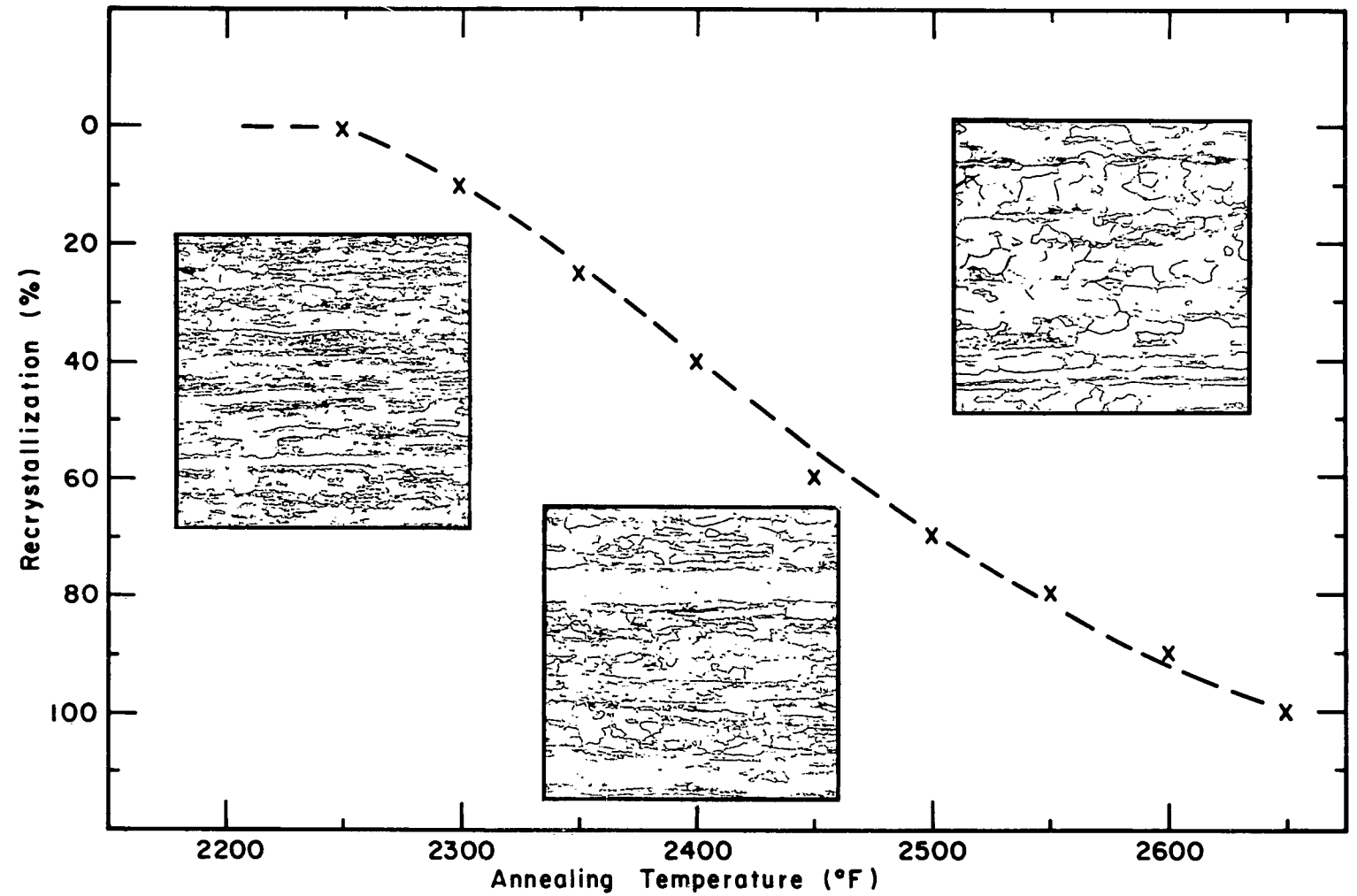
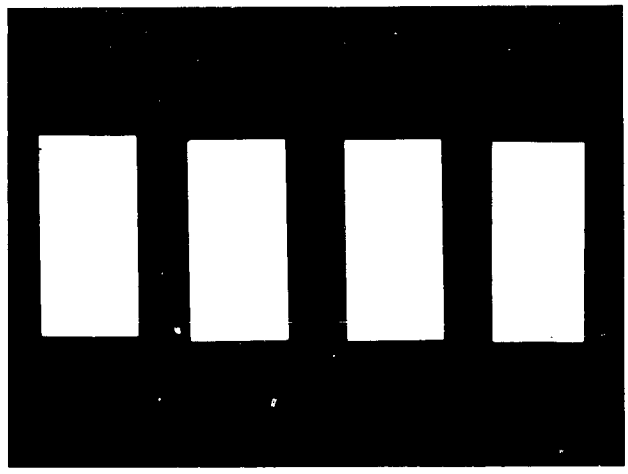


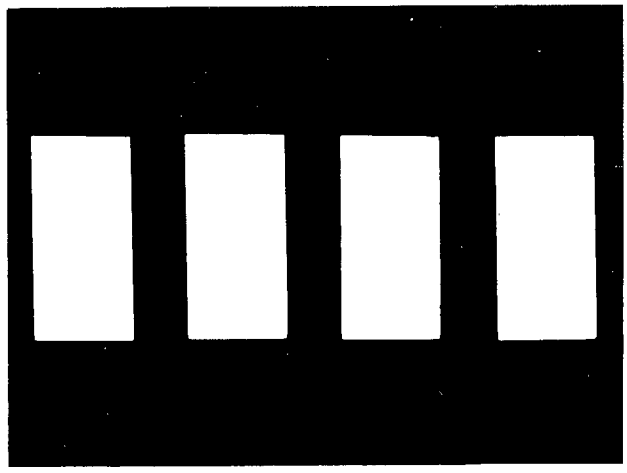
Figure 21

Recrystallization Behavior of Unalloyed Tungsten (1-Hour Annealing Time)



1420-62

75Ta-25W

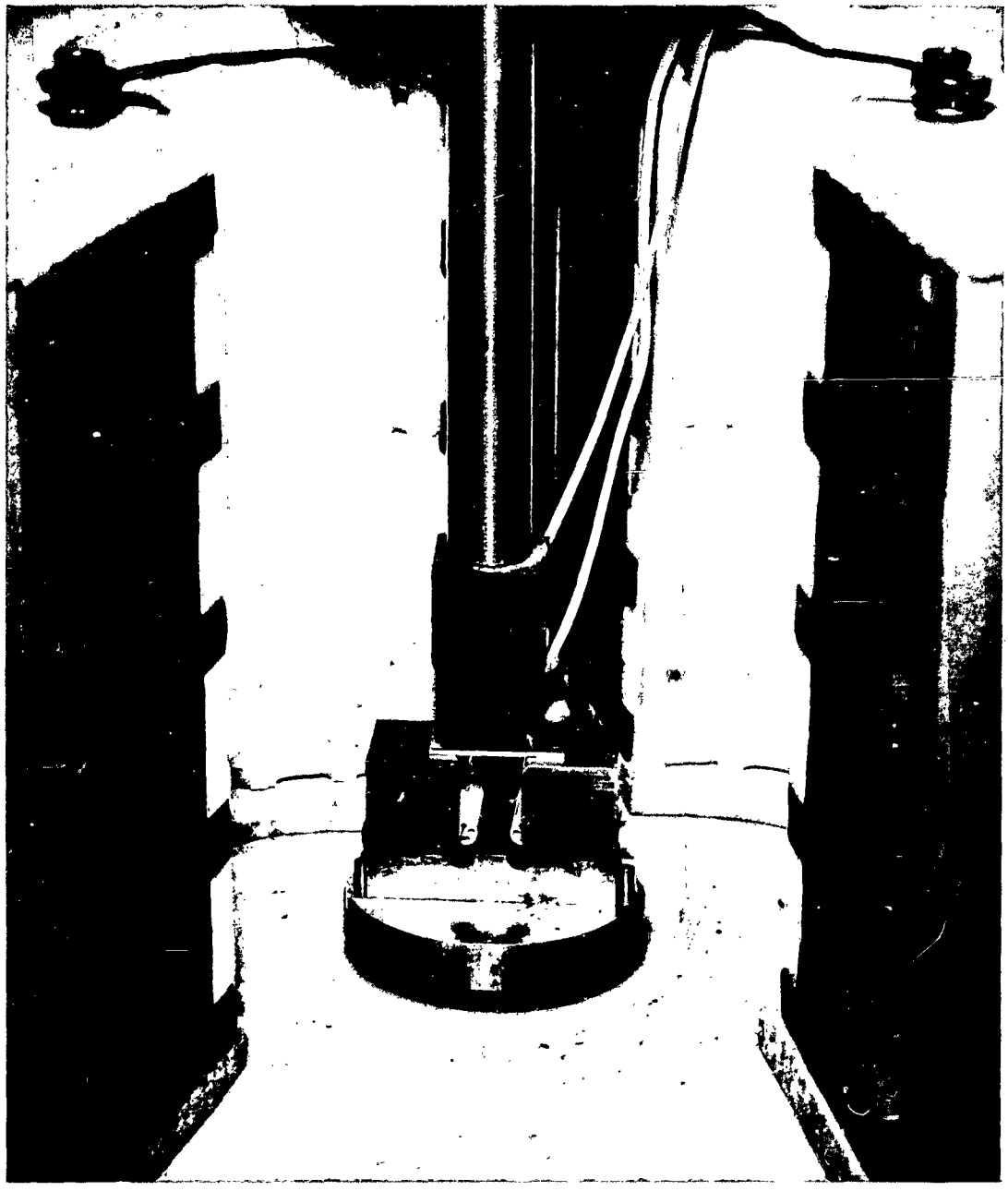


1426-62

88Ta-12Mo

Figure 22

Bend Test Specimens (Actual Size)



1500-62

Figure 23

Bend Test Equipment

Close-up of punch and die with bend specimen in position.

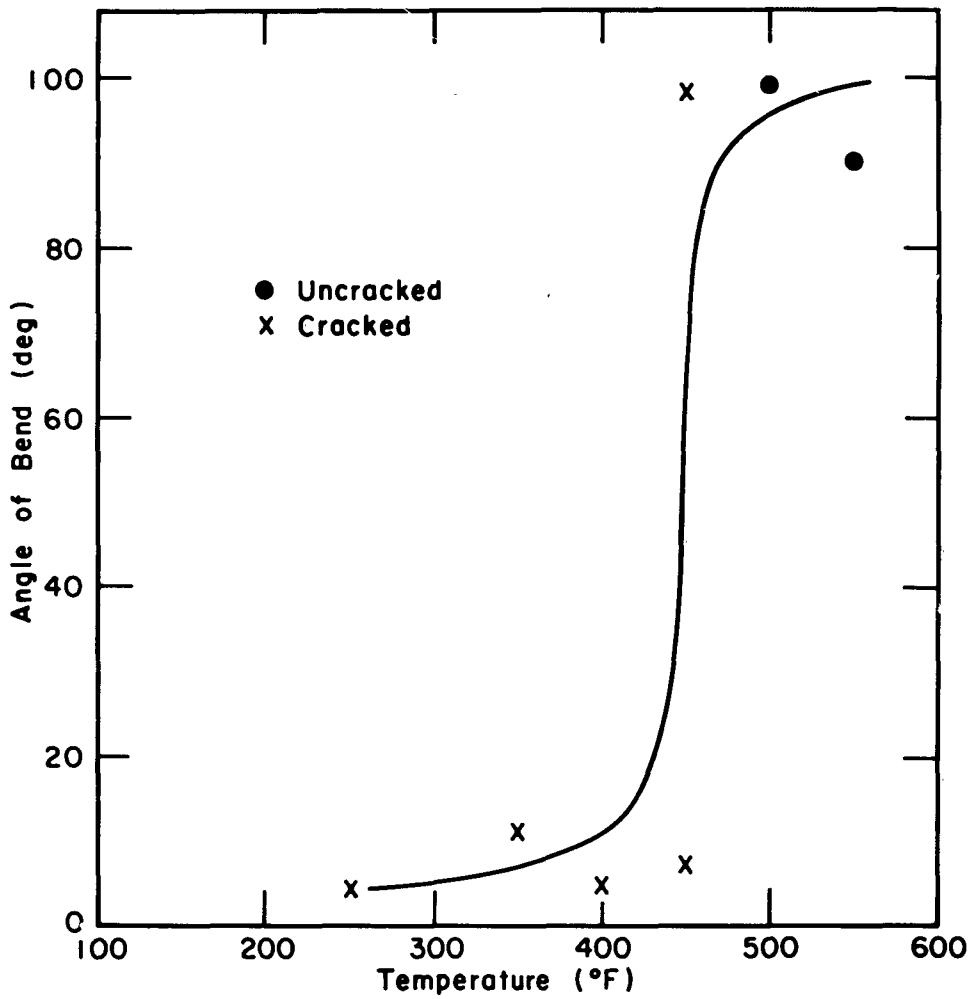


Figure 24

4T Bend Transition Curve for Unalloyed Tungsten Sheet after Stress Relief

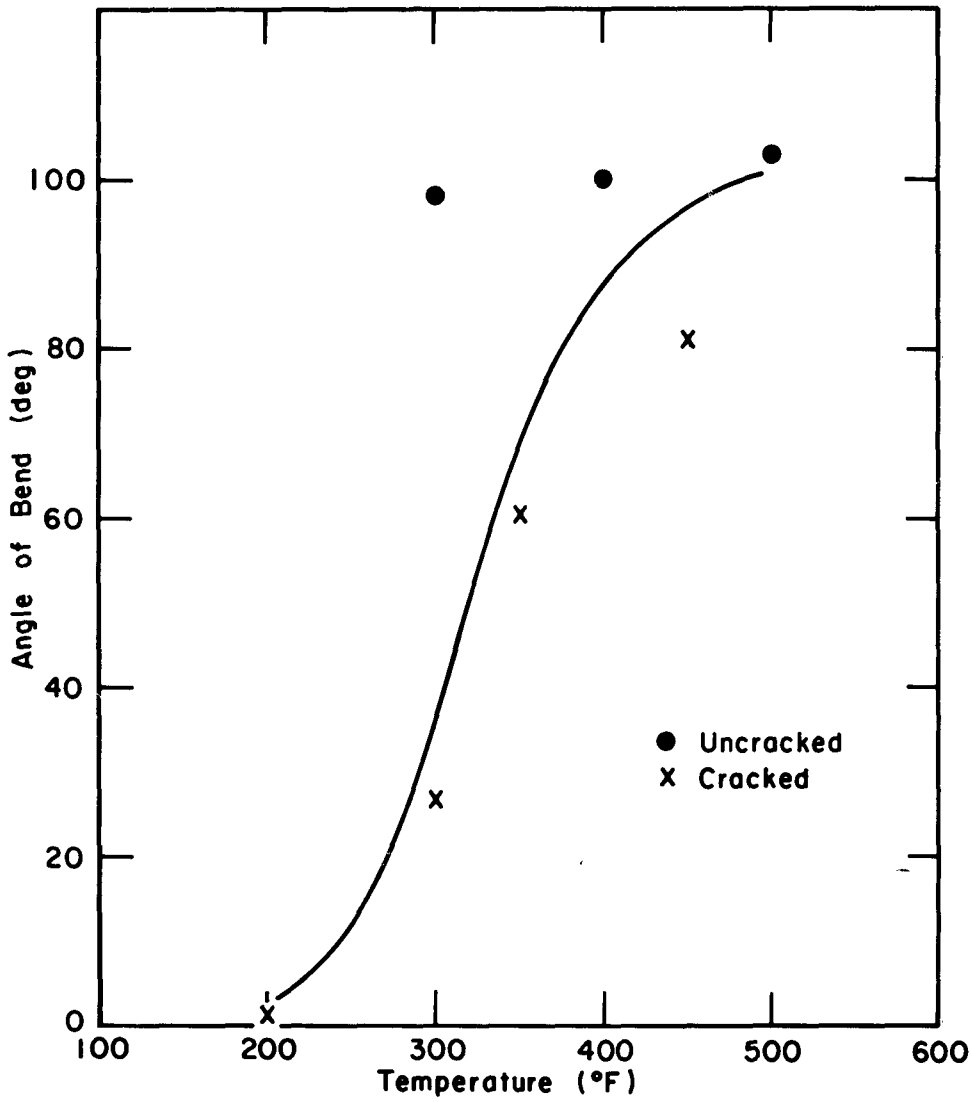


Figure 25

4T Bend Transition Curve for 88Ta-12Mo Sheet after Stress Relief

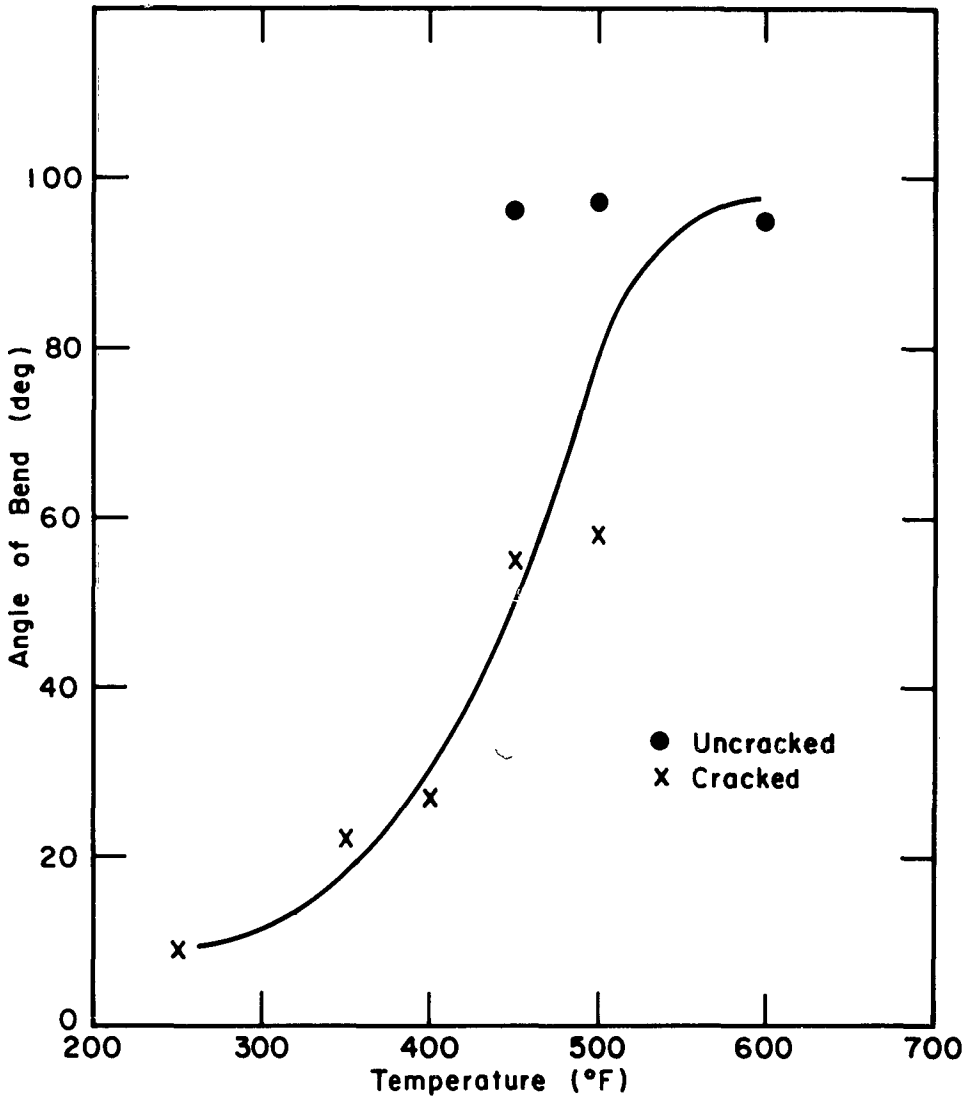


Figure 26

4T Bend Transition Curve for 75Ta-25W
Sheet after Stress Relief

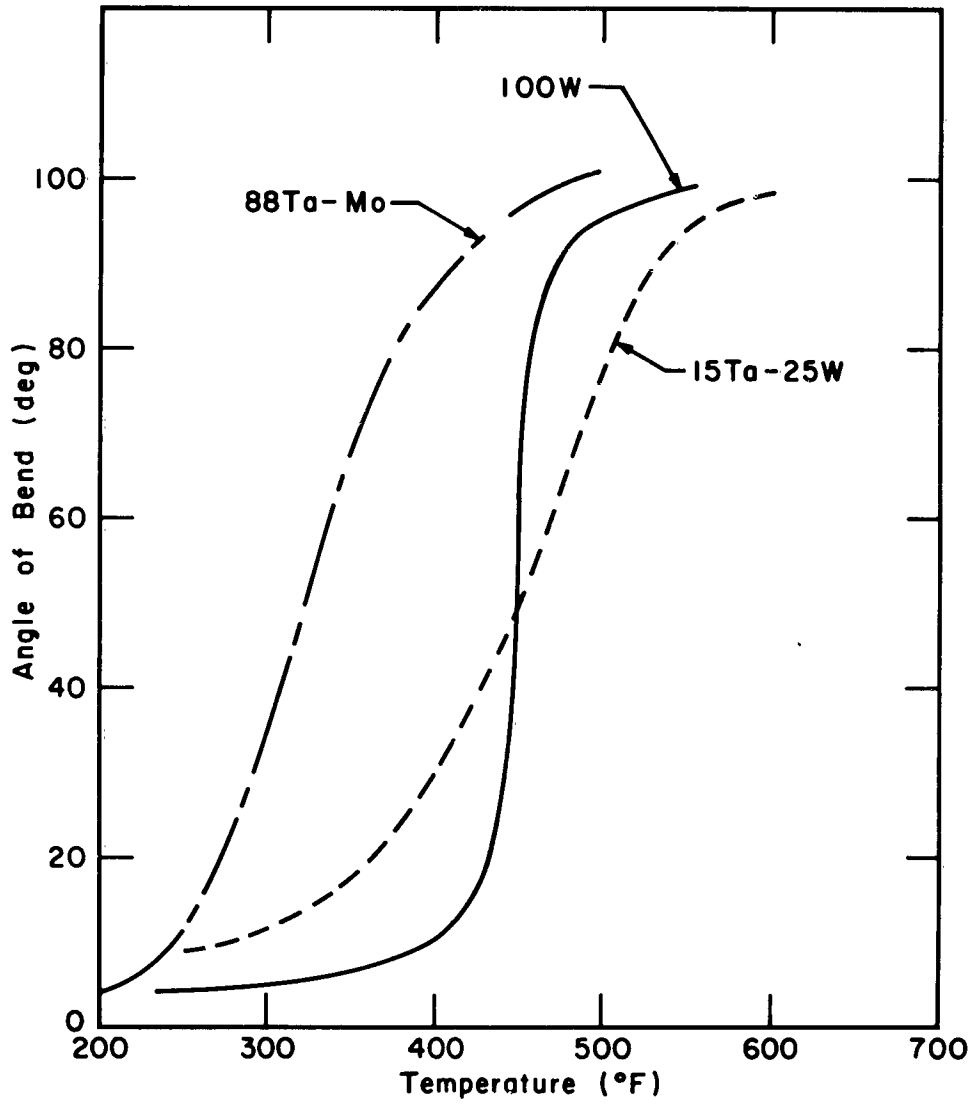
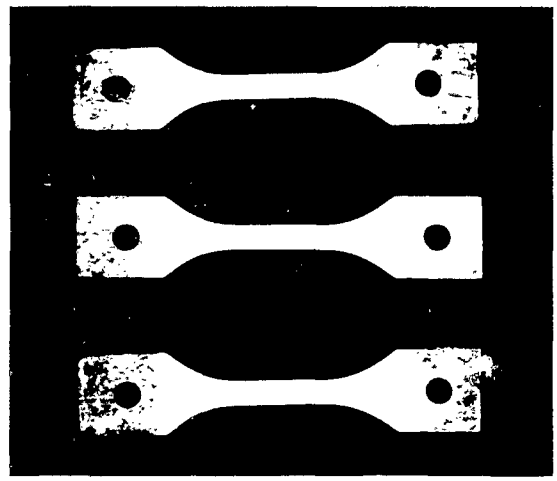


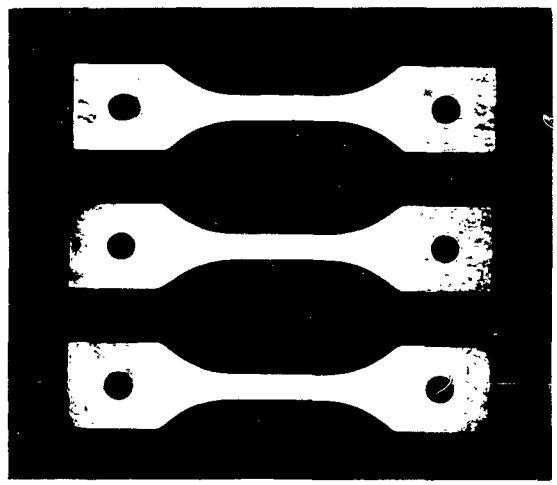
Figure 27

4T Bend Transition Curves for
Stress-Relieved Sheet



1420-62

75Ta-25W

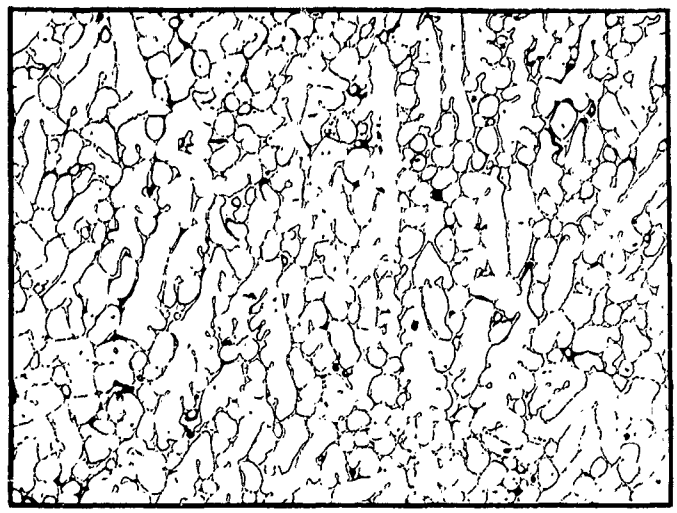


1426-62

88Ta-12Mo

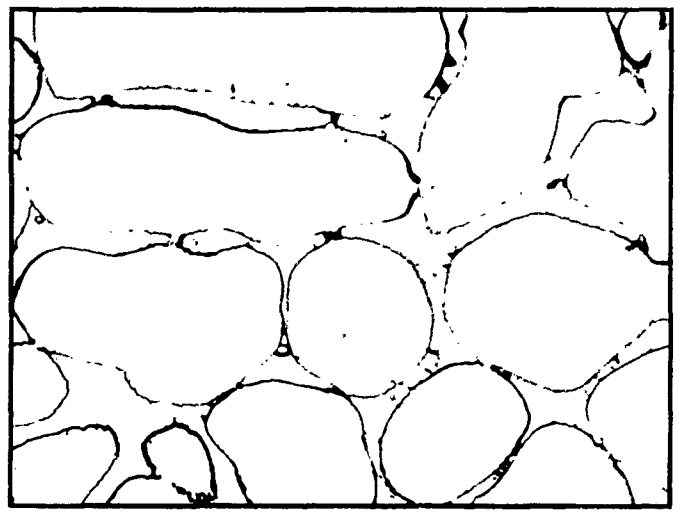
Figure 28

Microtensile Test Specimens (Actual Size)



170-62

(a) 100X



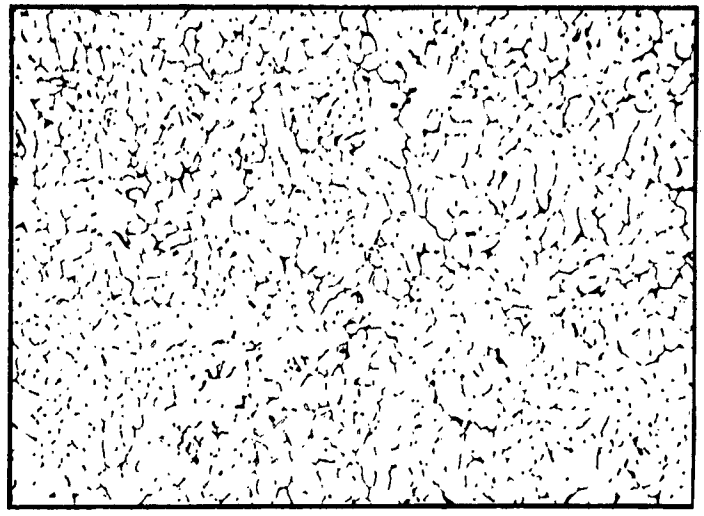
167-62

(b) 1000X

Figure 29

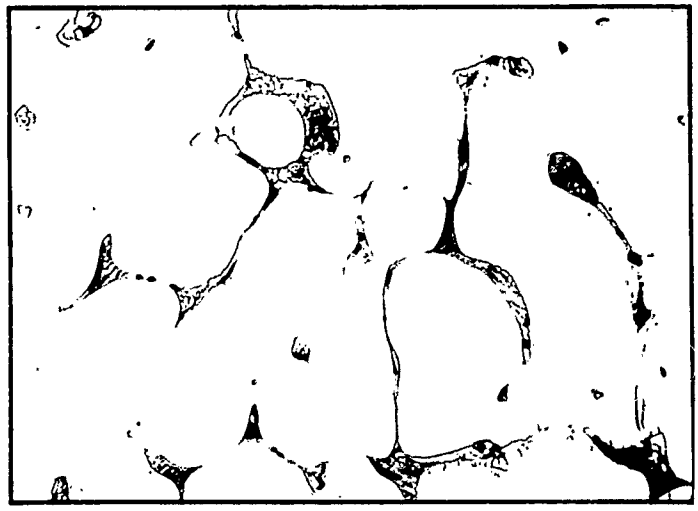
As-Cast Microstructure of the W-Ti-Zr-C Alloy (CEVA Heat 205)

Etchant: 50 cc Lactic Acid; 10 cc HNO_3; 1 cc HF



168-62

(a) 100X



165-62

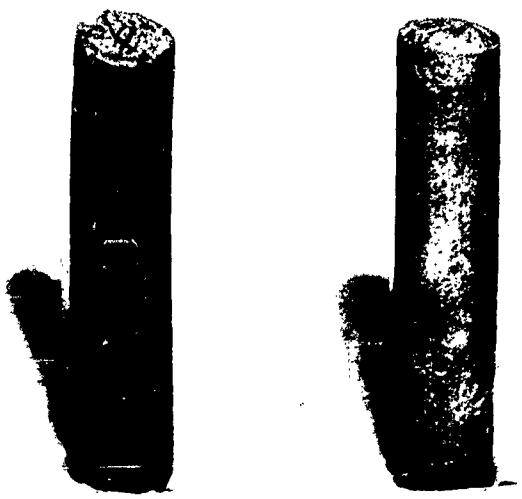
(b) 1000X

Figure 30

As-Cast Microstructure of the W-Cb-Zr-C Alloy (CEVA Heat 206)

Etchant: 50 cc Lactic Acid; 10 cc HNO_3; 1 cc HF

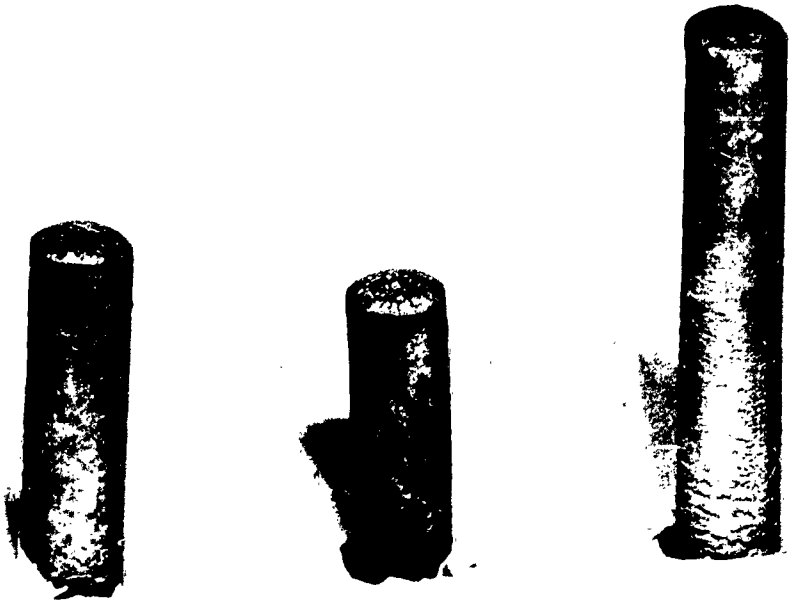
798-62



Heat 218B Heat 218T

Figure 31

Round-Bar Extrusions of the 88W-12Cb Alloy
(Actual Size)



799-62

Heat 207T Heat 207B Heat 229

Figure 32

Round-Bar Extrusions of the Phase II Alloys
(Actual Size)

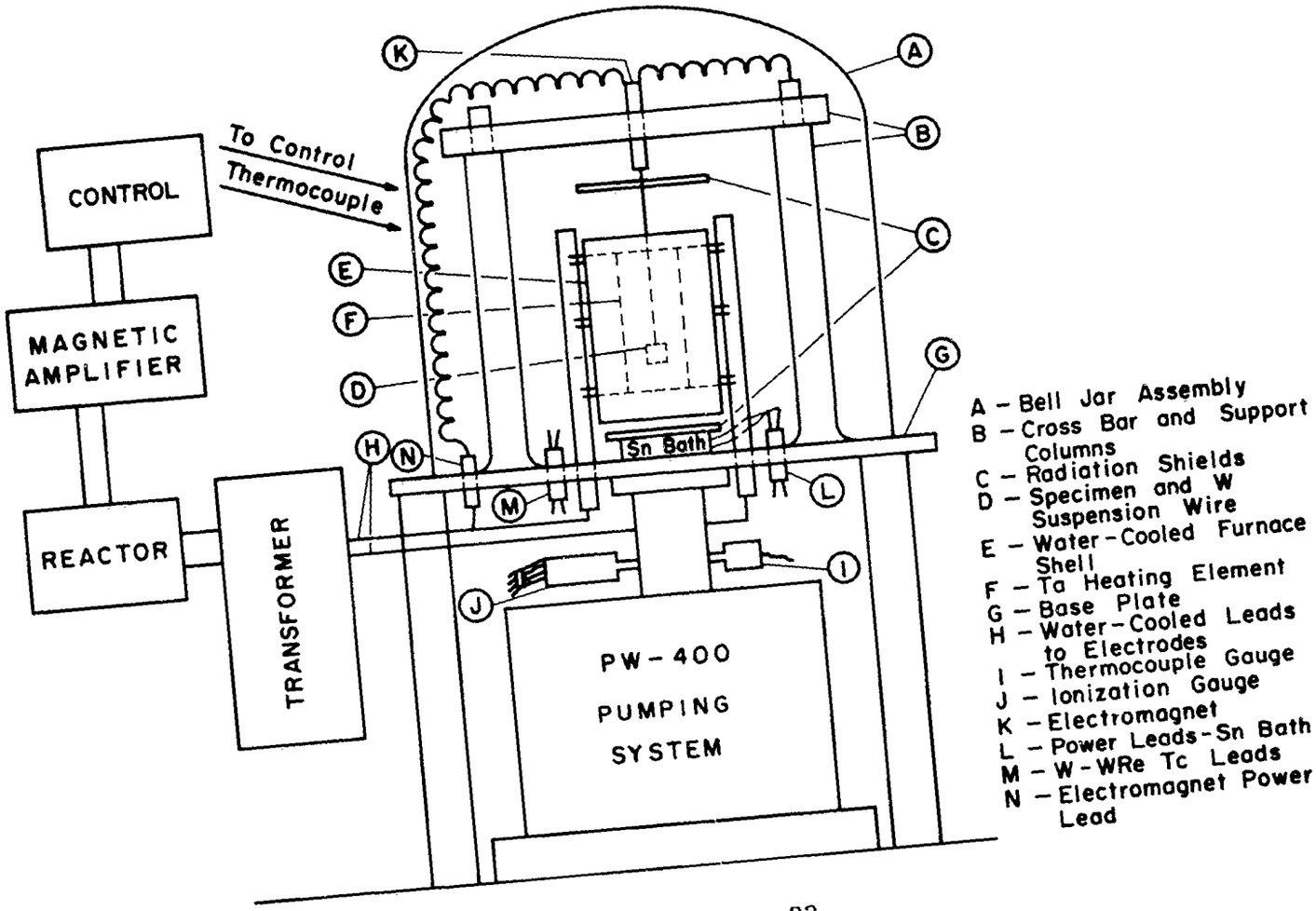
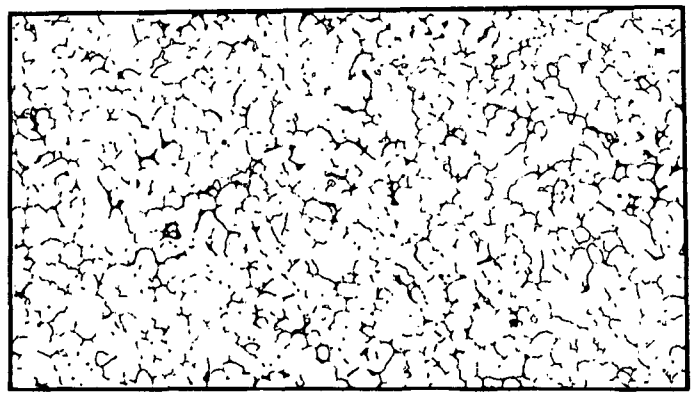


Figure 33

Schematic Drawing of Vacuum Heat-Treating Apparatus

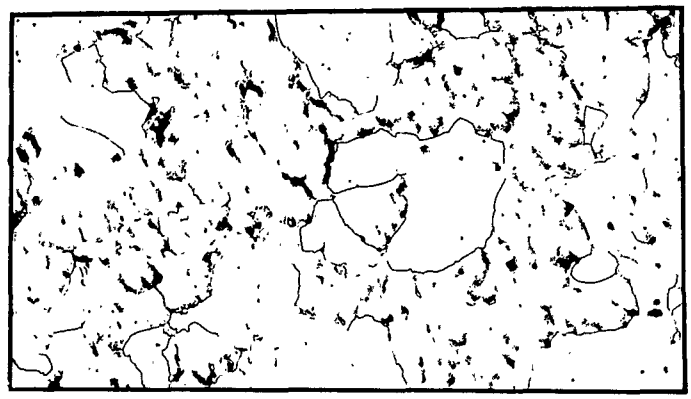
88W-12Cb Heat 218B

1662-62



W-12Cb-Zr-C Heat 207T

1651-62



W-12Cb-V-Zr-C Heat 229

1650-62

Figure 34

As-Cast Microstructures of Phase II Alloys (100X)

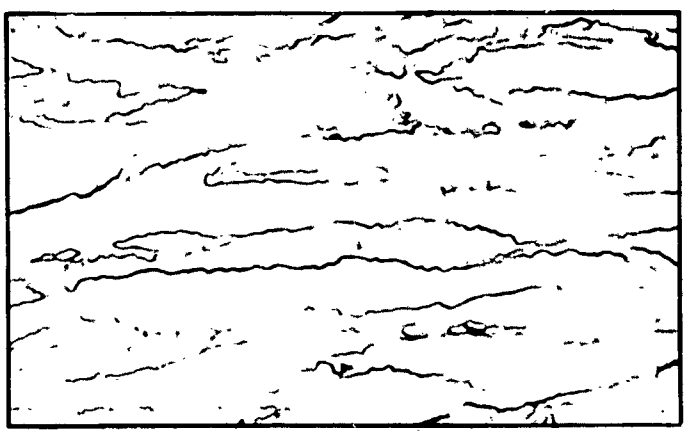
Etchant: Mixed Acids

484 DPH

As-Extruded

100X

1064-62

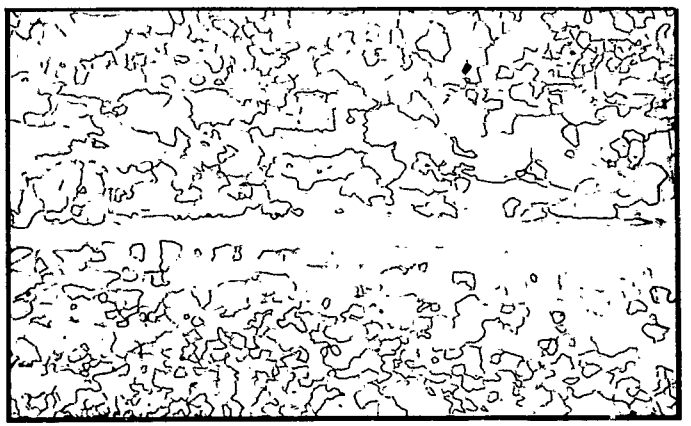


1000X

1130-62

395 DPH

3500 F for 1 hour, tin bath quenched



100X

1063-62

1000X

1135-62

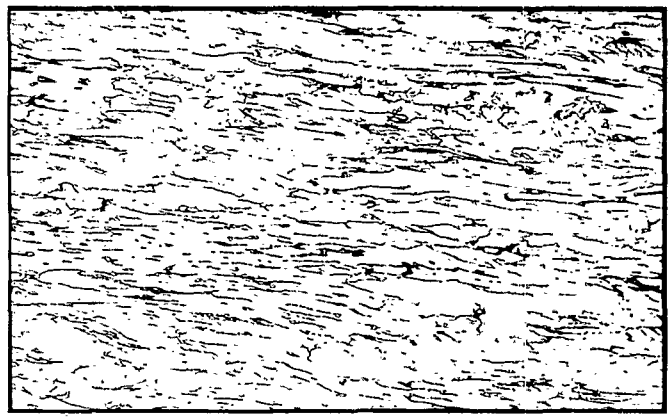
Figure 35

As-Extruded and Solution-Annealed Microstructures of the 88W-12Cb Alloy (Heat 218B)

Etchant: Mixed Acids

570 DPH

As-Extruded

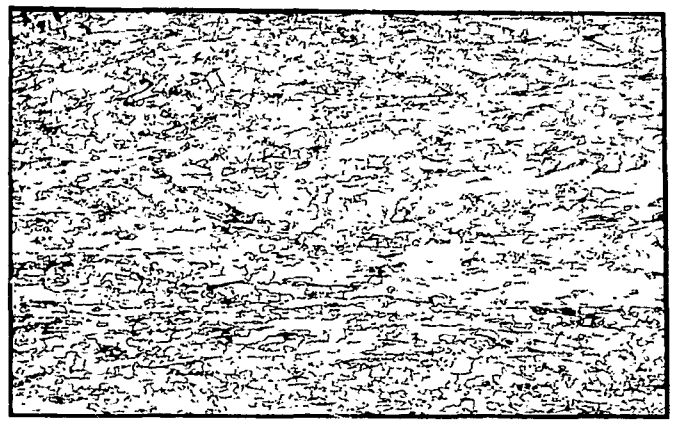


1060-62

100X

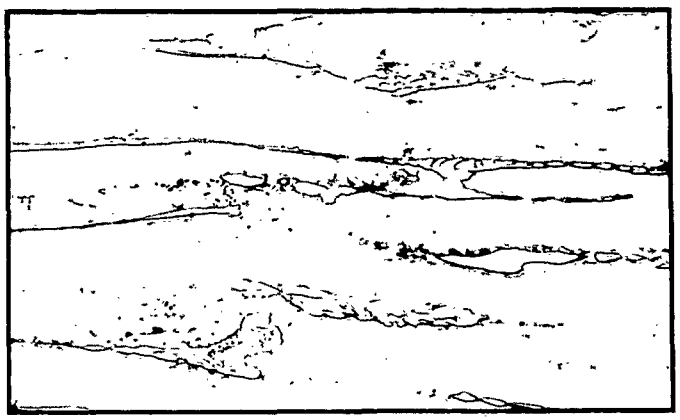
507 DPH

3500 F for 1 hour, tin bath quenched



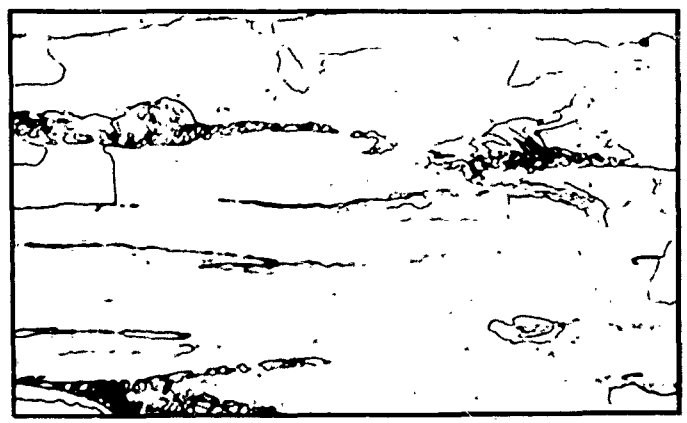
1062-62

100X



1121-62

1000X



1137-62

1000X

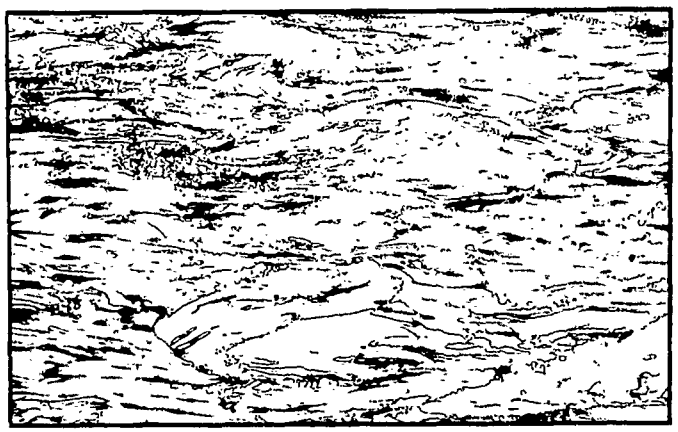
Figure 36

As-Extruded and Solution-Annealed Microstructures of the W-12Cb-Zr-C Alloy (Heat 207B)

Etchant: Mixed Acids

507 DPH

As-Extruded

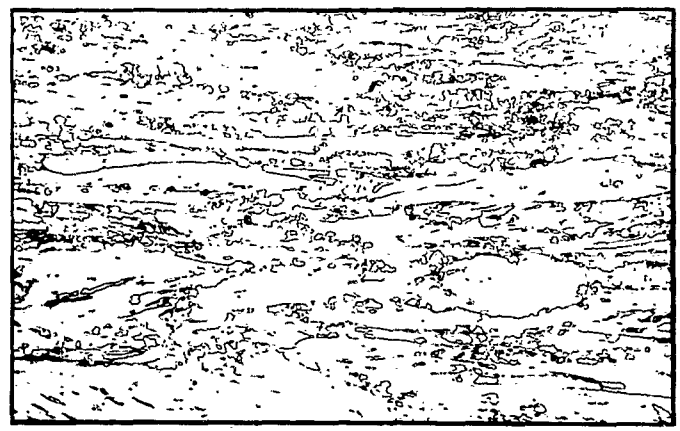


1051-62

100X

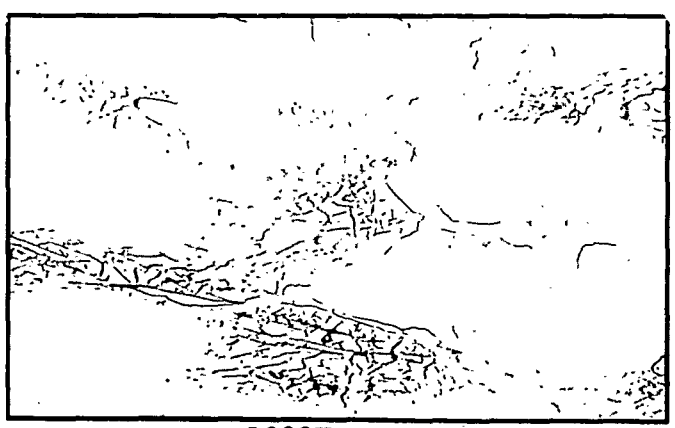
472 DPH

3500 F for 1 hour, tin bath quenched



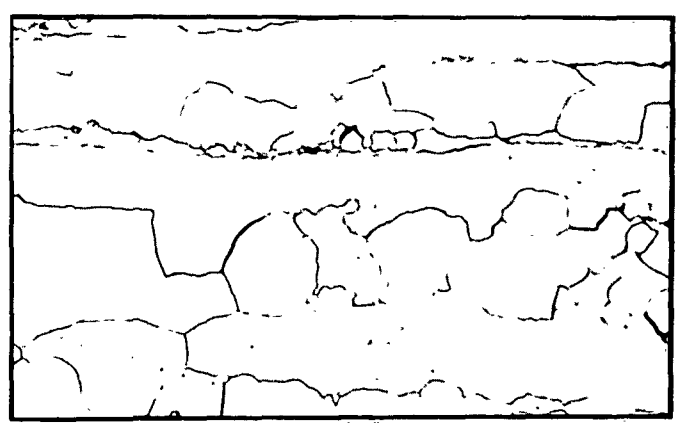
1053-62

100X



1125-62

1000X



1126-62

1000X

Figure 37

As-Extruded and Solution-Annealed Microstructures of the W-12Cb-V-Zr-C Alloy (Heat 229)

Etchant: Mixed Acids

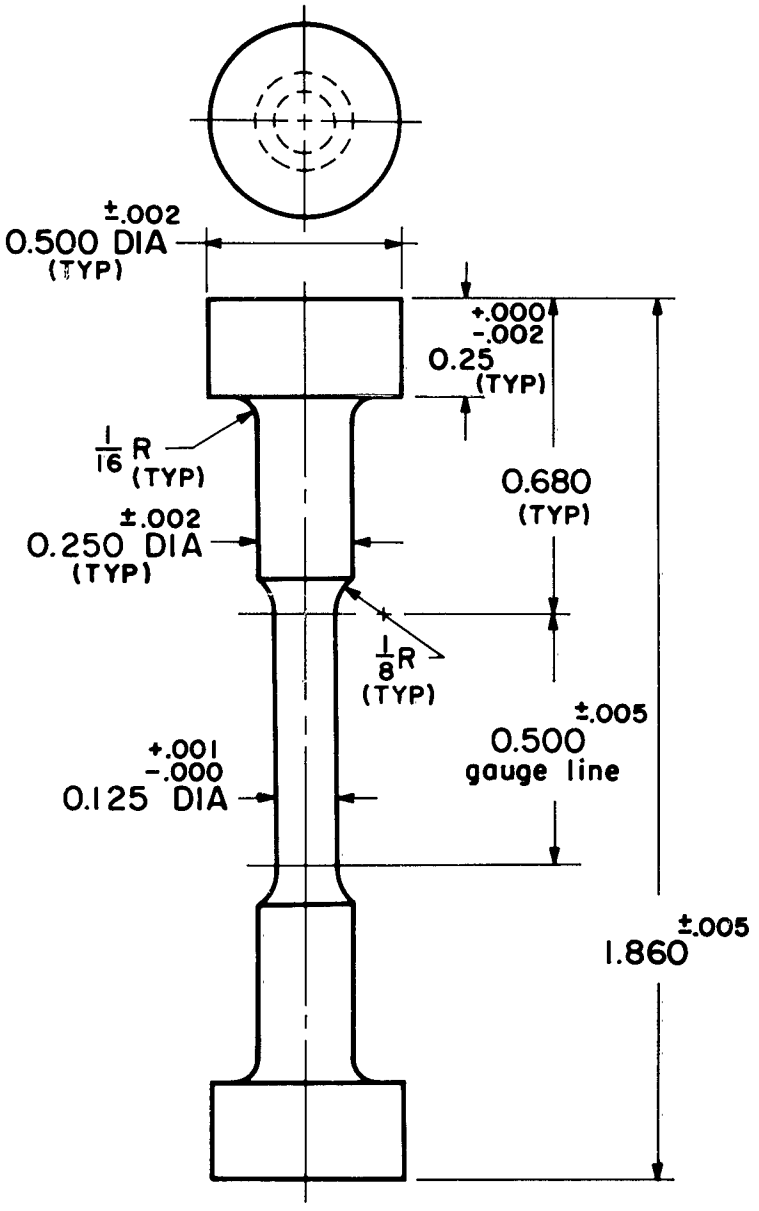
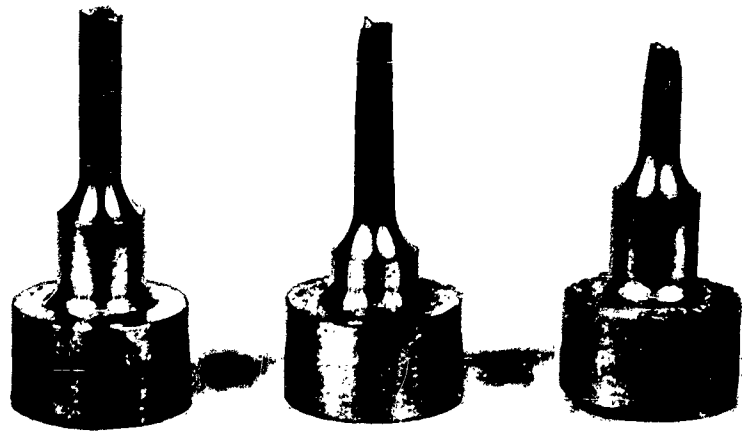


Figure 38

Tension Test Specimen

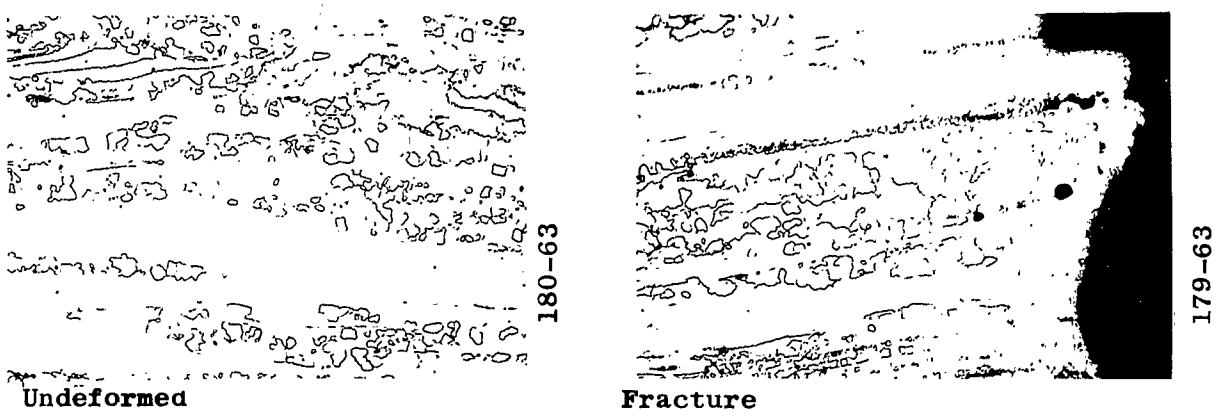


Heat 218B Heat 207T Heat 229

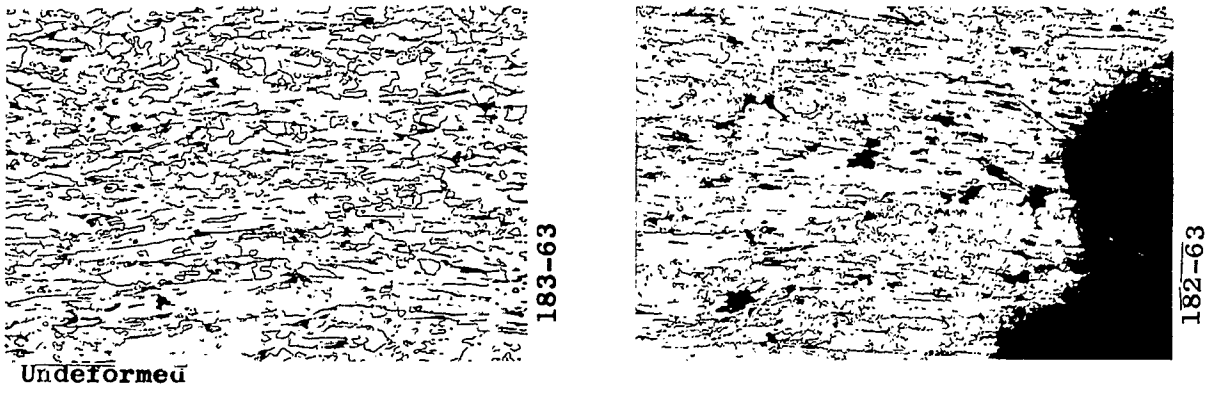
Figure 39

Tension Specimens Tested at 3500 F (2X)

Heat 218B: 88W-12Cb
Heat 207T: W-12Cb-Zr-C
Heat 229: W-12Cb-V-Zr-C



88W-12Cb (Heat 218B)



W-12Cb-Zr-C (Heat 207T)

W-12Cb-V-Zr-C (Heat 229)

Figure 40

Microstructures of Undeformed and Fractured Portions of Tensile Specimens (100X)

Etchant: Mixed Acids

Aeronautical Systems Division, Dir/Materials and Processes, Metals and Ceramics Lab, Wright-Patterson AFB, Ohio.
Rpt No. WADD-TR-61-134, Pt. II. RESEARCH ON WORKABLE REFRACTORY ALLOYS OF TUNGSTEN, TANTALUM, MOLYBDENUM, AND COLUMBIUM. Final report, Apr 63, 98p. incl illus., tables, 15 refs.

Unclassified Report

Under a previous contract, the W-Ta-Mo-Cb alloy system was investigated, and several tungsten- and tantalum-rich alloys were developed and evaluated in the form of extruded bars. Many of these alloys exhibited very high strengths at 3000 F; in fact, the tensile strengths of some alloys were in excess of 60,000 psi. The present work was a continuation of this effort and was aimed at producing and evaluating alloys from the W-Ta-Mo-Cb system in the form of sheet (Phase I) and increasing the high-

(over)

1. Refractory alloys
2. High temperature research

I. AFSC Project 7351, Task 735101
II. Contract AF 33(616)-8135
III. Crucible Steel Co. of America, Pittsburgh, Pa.
IV. R. C. Westgren, et al.
V. Aval fr OTS
VI. In ASTIA collection

Aeronautical Systems Division, Dir/Materials and Processes, Metals and Ceramics Lab, Wright-Patterson AFB, Ohio.
Rpt No. WADD-TR-61-134, Pt. II. RESEARCH ON WORKABLE REFRACTORY ALLOYS OF TUNGSTEN, TANTALUM, MOLYBDENUM, AND COLUMBIUM. Final report, Apr 63, 98p. incl illus., tables, 15 refs.

Unclassified Report

Under a previous contract, the W-Ta-Mo-Cb alloy system was investigated, and several tungsten- and tantalum-rich alloys were developed and evaluated in the form of extruded bars. Many of these alloys exhibited very high strengths at 3000 F; in fact, the tensile strengths of some alloys were in excess of 60,000 psi. The present work was a continuation of this effort and was aimed at producing and evaluating alloys from the W-Ta-Mo-Cb system in the form of sheet (Phase I) and increasing the high-

(over)

1. Refractory alloys
2. High temperature research

I. AFSC Project 7351, Task 735101
II. Contract AF 33(616)-8135
III. Crucible Steel Co. of America, Pittsburgh, Pa.
IV. R. C. Westgren, et al.
V. Aval fr OTS
VI. In ASTIA collection

temperature strength of alloys of this type by the formation of dispersed carbides (Phase II). For Phase I, small cylindrical ingots of six selected alloys and unalloyed tungsten were consumably vacuum arc-melted by a multiple electrode technique and successfully extruded to sheet bars. Two alloys and unalloyed tungsten were rolled to sheet and recrystallization temperatures, bend transition temperatures, and high-temperature tensile properties were determined. The other four alloys could not be rolled to sheet by the techniques attempted in this program. Experiments under Phase II resulted in the development of a successful technique for consumably vacuum arc melting 88W-12Cb alloys that contain small amounts of vanadium, zirconium, and carbon. In the as-extruded condition, the alloys showed 3500 F tensile strengths of 49,000 to 57,000 psi — this constituted a twofold increase in strength above that of the base solid solution composition.

temperature strength of alloys of this type by the formation of dispersed carbides (Phase II). For Phase I, small cylindrical ingots of six selected alloys and unalloyed tungsten were consumably vacuum arc-melted by a multiple electrode technique and successfully extruded to sheet bars. Two alloys and unalloyed tungsten were rolled to sheet and recrystallization temperatures, bend transition temperatures, and high-temperature tensile properties were determined. The other four alloys could not be rolled to sheet by the techniques attempted in this program. Experiments under Phase II resulted in the development of a successful technique for consumably vacuum arc melting 88W-12Cb alloys that contain small amounts of vanadium, zirconium, and carbon. In the as-extruded condition, the alloys showed 3500 F tensile strengths of 49,000 to 57,000 psi — this constituted a twofold increase in strength above that of the base solid solution composition.